Symmetry and Magnetic Structures

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Introduction

The crystallographic approach to magnetic structures is largely based on two kinds of descriptions: symmetry invariance of magnetic configurations (Magnetic Space Groups, often called Shubnikov groups) and group representation theory applied to conventional crystallographic space groups. The first approach is nearly exclusively used for the case of commensurate magnetic structures and usually is limited to a description of the invariance symmetry properties for this kind of configurations [1-5]. The representation analysis is more general and can be applied to all kinds of magnetic structures. The literature on this field is broad and one can consult the papers of E.F. Bertaut [6-9] and Y. Izyumov and co-workers [10-15] to get a deeper insight into the problem. A possible extension of the Shubnikov approach may be developed in the sense of Magnetic Superspace Groups as in crystallographic incommensurate structures. This more general approach extends the invariance concept to incommensurate magnetic structures and may be a better approach when macroscopic properties have to be deduced from the spin configurations. At present not too much work has been done in that sense so it will not be considered in the present notes.

The outline of this document is as follows:

1. What is a magnetic structure?

The appearance of magnetic moments in atoms is due to the existence of unpaired electrons in some electronic shells. The Hund's rule favours the "ferromagnetic" interaction between electrons inside the same ion when they are in different orbitals, so that an intrinsic moment appears in such cases. The intrinsic moment contains, in general, an orbital and a spin contribution. For our purposes we assume that there are atoms, in positions $\mathbf{R}$, with magnetic moments that are usually disordered in the paramagnetic state ($\forall \mathbf{R}, \langle \mathbf{m_R} \rangle_t = 0$, where the bracket symbol means time average) and become frozen ($\langle \mathbf{m_R} \rangle_t \neq 0$), and eventually ordered, below a certain temperature. A magnetic structure corresponds to a particular, nearly static, spatial arrangement of magnetic moments that sets in below the ordering temperature. Above the ordering temperature the system is in the paramagnetic state. In this document we disregard the quantum properties of spin operators and we interchange loosely the terms magnetic moment (generated by the spin and orbital angular momentum of electrons) and "atomic total spin" or "spin" for short. For us, the magnetic moment of an atom is a classical "axial vector" (magnetic dipole) supposed to be generated by an "electrical current".

The magnetic structures are commonly represented as a set of arrows, associated to the magnetic atoms, with magnitudes and orientations characteristics of the particular magnetic structure. For considering symmetry properties we have to suppose that each arrow is in fact a current loop, the direction of which determines the orientation of the arrow by the right-hand rule of electromagnetism.

What governs the setting up of a particular kind of magnetic ordering? Let us consider a classical and general Hamiltonian of interacting spins (in fact magnetic moments) of the following form:

$$H = -\sum_{il\alpha, jm\beta} J_{il,jm}^{\alpha\beta} S_{il\alpha} S_{jm\beta} + \sum_{il\alpha, jm\beta, kn\gamma} J_{il,jm,kn}^{\alpha\beta\gamma} S_{il\alpha} S_{jm\beta} S_{kn\gamma} + ... O(S^n) \qquad (1)$$

Where the indices i, j, k label atoms in the primitive cell, the indices l, m, n label different unit cells (they are composite indices, describing the origin of the unit cells at lattice vectors $\mathbf{R}_l$, $\mathbf{R}_m$ and $\mathbf{R}_n$) and the indices α, β, γ label the components x, y, z. If we consider that the magnetic atoms at positions $\mathbf{R}_{lj}$ have intrinsic magnetic moments of module m_j ($\mathbf{S}_{lj}=m_j \mathbf{n}_{lj}$), for a given set of exchange parameters $\{J_{il,jm}^{\alpha\beta}, J_{il,jm,kn}^{\alpha\beta\gamma}, ...\}$, the magnetic structure at $T = 0$ K corresponds to the minimum of H with respect to the orientations of the unit vectors $\mathbf{n}_{lj}$. In general the terms of order greater than 2 in the spins are much weaker than those of second order. Moreover the isotropic part of the tensors $J_{il,jm}^{\alpha\beta}$ is strongly dominant in many cases; the energy can be simplified to obtain the so called Heisenberg Hamiltonian:

$$H_{iso} = -\sum_{il, jm} J_{il,jm} \mathbf{S}_{il} \cdot \mathbf{S}_{jm} = -\sum_{il, jm} J_{ij}(\mathbf{R}_m\text{-}\mathbf{R}_l) \, \mathbf{S}_{il} \cdot \mathbf{S}_{jm}$$

$$H_{iso} = -\sum_{il, jm} J_{ij}(\mathbf{R}_m\text{-}\mathbf{R}_l) \, m_i m_j \, \mathbf{n}_{il} \cdot \mathbf{n}_{jm} = -\sum_{il, jm} J_{ij}(\mathbf{R}_m\text{-}\mathbf{R}_l) \, m_i m_j \cos\theta_{il, jm} \qquad (2)$$

The magnetic energy in such a case does not depend on the absolute orientation of the spins. It depends only on the relative orientation of the spins (scalar product). This means that for a given configuration, any global rotation acting on the spins does not change the energy, so the Hamiltonian has more symmetry elements than the space group underlying the crystal structure if the interaction between the magnetic moments is strictly isotropic. We see here that the relevant symmetry to be considered for studying the magnetic ordering is that of the

Hamiltonian of the system. However the Hamiltonian of a system is generally unknown, so we do not know what is the relevant symmetry to begin with. In the absence of this information we have at our hands the possibility to know the symmetry of the crystal structure of the compound under study. The space group of the crystal summarises in some way the result of the different interactions between electrons and nuclei in the solid. We expect that the magnetic ordering conserves some of the symmetry properties of the crystallographic structure even in the case of a structural transition accompanying the magnetic ordering. This is the reason why we use the symmetry of the crystal structure as a starting point for our analysis. In the course of the analysis of experimental data we may arrive to the conclusion that we have to modify our previous assumptions on the starting symmetry; or even to suppose that we have more symmetry (additional symmetry elements) in the magnetic Hamiltonian than in the crystallographic space group.

2. Invariance properties of magnetic structures.

We assume that the reader is familiar with crystallography and with the crystallographic point and space groups (see chapter by Grenier & Ballou). The symmetry operators used in crystal structures can also be used to study the invariance properties of magnetic structures: We have then to consider, in addition to positions of point atoms, a property associated to each point atom that is represented by an axial vector (an arrow surrounded by a current loop). To study the invariance of magnetic configurations we have to introduce a new operator that is usually called "spin reversal" or "time reversal". This operator is acting on magnetic moments that are "classical axial vectors"[1]. The action consists of changing the sense of the current loop (proportional to the product of a charge by a velocity vector), so that the orientation of the magnetic moment is reversed. We note this operator as $1'$ and it acts only on the magnetic moments/spins not on the atom positions: $1' \cdot \mathbf{m} = -\mathbf{m}$.

Figure 1: Action of time reversal in a current loop: the magnetic dipole is inverted
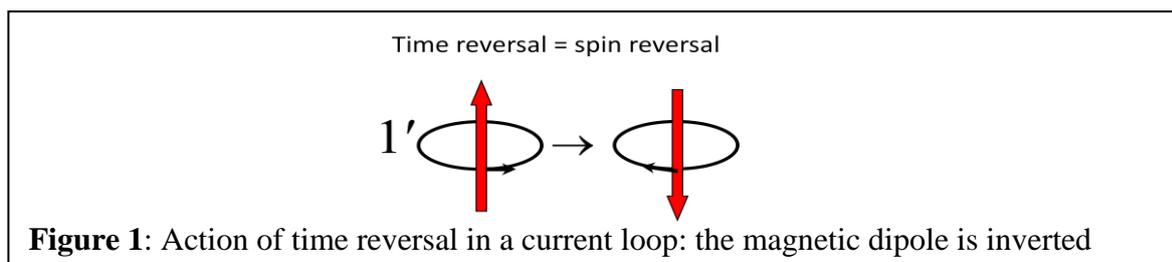
The spin reversal operator cannot be contained in the set of symmetry operators that leave invariant a magnetically ordered system, however it is contained in the paramagnetic state of a magnetic system. This gives immediately the so called "paramagnetic space groups" that are obtained by adding $1'$ to the set of symmetry elements of the space group. The notation of the paramagnetic space groups is identical to that of crystallographic groups with the $1'$ symbol added (e.g. Pbnm1'). In magnetically ordered systems the spin reversal operator can be combined with a conventional symmetry operator. The operator is called "primed" to indicate that we have to invert the spin after applying the "non-primed" operator to the spin.

The symmetry operators we have to consider for exploring the invariance of spin configurations are then formed by the usual operators considered in crystallography together with these same operators followed by the spin reversal.

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[1] In quantum mechanics "time reversal" is an anti-unitary operator acting on wave functions and quantum operators and is related to "conjugation" for spinless systems. We will not treat this point in this document.

2.1: *Behaviour of axial vectors under the action of operators representing general motions*

Axial vectors are in fact second order antisymmetric tensors in the conventional 3D Euclidean space. The three independent components of these tensors can be written as components of a vector with the particularity, contrary to *polar* vectors, that the orientation of the vector depends on the handedness of the reference frame. Examples of polar vectors are, for instance, the vector position, velocity and acceleration of a particle, the electrostatic field, etc. The behaviour of axial vectors under the action of motions (translations, proper and improper rotations, glide planes and screw axes) is different from that of polar vectors. This can be visualised in the following scheme representing the action of a mirror plane on an electric dipole and on a magnetic dipole (magnetic moment).

Figure 2: Action of a mirror plane on an electrical dipole (polar vector) and on a magnetic dipole (axial vector)
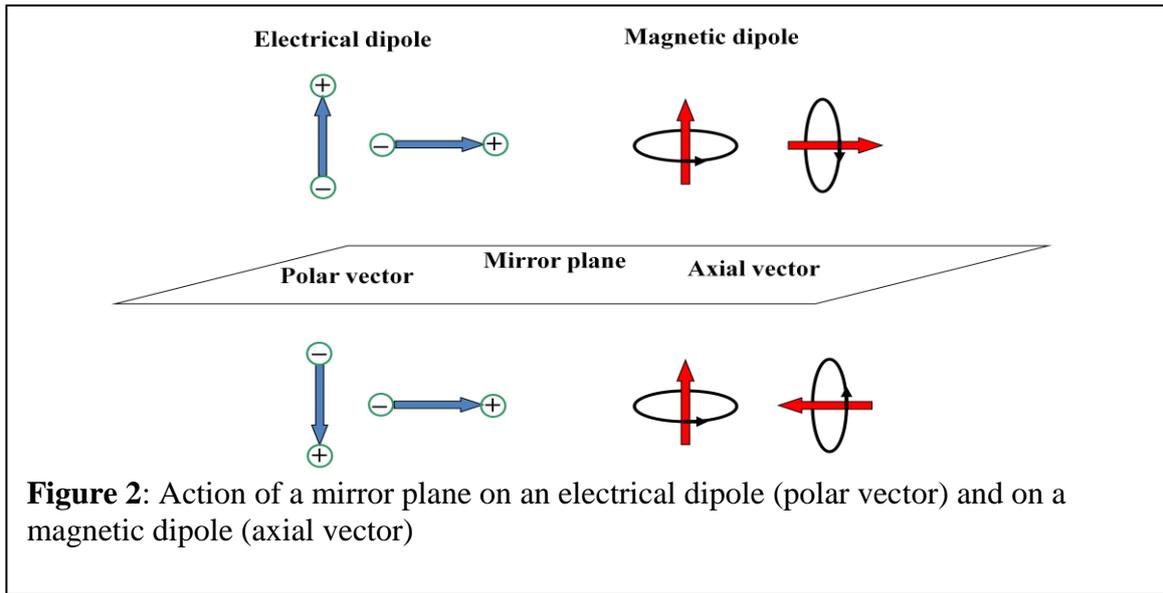
Mathematically the action of whatever kind of symmetry operator, which can be represented by an orthogonal matrix in the appropriate reference frame, on an axial vector is identical to that on a polar vector except that we have to multiply by the determinant of the matrix. Moreover if we consider a "primed" element we have to multiply again by -1 all the components of the resulting vector. In summary a general operator $g = \{h \,|\, \mathbf{t}_h + \mathbf{n}\}$ (see chapter by Grenier & Ballou) acting on an atom $\mathbf{r}_j$ in the cell at the origin (zero-cell) having a magnetic moment $\mathbf{m}_j$ is transformed as follows:

Position:
$$\mathbf{r}_j' = g\,\mathbf{r}_j = \{h \,|\, \mathbf{t}_h + \mathbf{n}\}\mathbf{r}_j = h\mathbf{r}_j + \mathbf{t}_h + \mathbf{n} = \mathbf{r}_i + \mathbf{a}_{gj} \tag{3}$$

Magnetic moment:
$$\mathbf{m}_j' = g\mathbf{m}_j = \det(h)\,\delta\,h\mathbf{m}_j \tag{4}$$

The transformed atom position $\mathbf{r}_j'$ is translationally equivalent to the atom labelled "i" in the zero-cell. The vector $\mathbf{a}_{gj}$ is a lattice vector depending on the initial atom j and the operator g. It is called "returning vector" because its opposite is the vector we need to apply on the transformed atom to put it back in the zero-cell.

The corresponding magnetic moment is transformed using only the rotational part of the operator and the resulting vector is multiplied by the determinant and the "signature" (δ) of the operator. The signature is $\delta=1$ for unprimed elements and $\delta = -1$ for primed elements.

In a magnetically ordered system the operator g is a symmetry operator if it is a symmetry operator of the space group and if $\mathbf{m}_j' = \mathbf{m}_i$.

2.2: *Magnetic point groups, Magnetic lattices and magnetic space groups*

Let us define the time reversal group that is formed by only two elements: $\mathbf{R} = \{1, 1'\}$. Whatever crystallographic magnetic group, $\mathbf{M}$, can be obtained as a subgroup of the exterior direct product of $\mathbf{R}$ by the crystallographic group $\mathbf{G}$: $\mathbf{M} \subset \mathbf{G} \otimes \mathbf{R}$. The group $\mathbf{G}$ is always a magnetic group (called "colourless"). The paramagnetic ("grey") groups of the form $\mathbf{P} = \mathbf{G} + \mathbf{G}1'$ are also magnetic groups. The fact that the product of two primed elements must be an unprimed element gives the fundamental lemma for constructing the rest of magnetic groups ("black-white" groups): *the magnetic groups derived from the crystallographic group* $\mathbf{G}$ *can be constructed considering the index 2 subgroups* $\mathbf{H}$ *of* $\mathbf{G}$ *as constituting the unprimed elements and the rest of operators*, $\mathbf{G} - \mathbf{H}$, *those that are multiplied by the time reversal operator*. Remember that the index of $\mathbf{H}$ in $\mathbf{G}$ is the ratio of the orders of $\mathbf{G}$ over $\mathbf{H}$, $[i](\mathbf{H}/\mathbf{G}) = n(\mathbf{G})/n(\mathbf{H})$. The magnetic group is then related to the subgroup $\mathbf{H} \subset \mathbf{G}$ (of index 2) by the expression:

$$\mathbf{M} = \mathbf{H} + (\mathbf{G} - \mathbf{H})\, 1'. \tag{5}$$

This is valid for all kind of groups: point groups, translation groups and space groups.

Even without tables of magnetic group one can construct easily different magnetic groups looking into the International Tables of Crystallography, Volume A (ITA), where the subgroups of index 2 for all the space groups are given.

Fig. 10.3.2. Maximal subgroups and minimal supergroups of the three-dimensional crystallographic point groups. Solid lines indicate maximal normal subgroups; double or triple solid lines mean that there are two or three maximal normal subgroups with the same symbol. Dashed lines refer to sets of maximal conjugate subgroups. The group orders are given on the left. Full Hermann–Mauguin symbols are used.

781

Figure 3 (reproduced from ITA)

Let us consider an example for magnetic point groups. One has to consult the scheme of the group/subgroup relations as given in page 781 of ITA and reproduced in Figure 3.

Consider the point group $\mathbf{G} = 4/m$ of order 8. Its subgroups of index 2 are $\mathbf{H}_1 = 4$, $\mathbf{H}_2 = \bar{4}$ and $\mathbf{H}_3 = 2/m$:

$$\mathbf{G} = 4\,/\,m = \{1, 4_z^+, 2_z, 4_z^-, \bar{1}, m_z, \overline{4_z^+}, \overline{4_z^-}\}$$

$$\mathbf{H}_1 = 4 = \{1, 4_z^+, 2_z, 4_z^-\}$$

$$\mathbf{H}_2 = \bar{4} = \{1, \overline{4_z^+}, 2_z, \overline{4_z^-}\}$$

$$\mathbf{H}_3 = 2\,/\,m = \{1, 2_z, \bar{1}, m_z\}$$

There are then four magnetic point groups derived from **G**, they have the following elements:

$$\mathbf{M}_0 = \mathbf{G} = 4\,/\,m$$

$$\mathbf{M}_1 = \mathbf{H}_1 + (\mathbf{G} - \mathbf{H}_1)1' = \{1, 4_z^+, 2_z, 4_z^-, \bar{1}', m_z', \overline{4_z^+}', \overline{4_z^-}'\} = 4\,/\,m'$$

$$\mathbf{M}_2 = \mathbf{H}_2 + (\mathbf{G} - \mathbf{H}_2)1' = \{1, 4_z^+', 2_z, 4_z^-', \bar{1}', m_z', \overline{4_z^+}, \overline{4_z^-}\} = 4'/\,m'$$

$$\mathbf{M}_3 = \mathbf{H}_3 + (\mathbf{G} - \mathbf{H}_3)1' = \{1, 4_z^+', 2_z, 4_z^-', \bar{1}, m_z, \overline{4_z^+}', \overline{4_z^-}'\} = 4'/\,m$$

2.3: *Magnetic point groups*

Following the previous procedure one can construct the full list of magnetic point groups. It is easy to apply the lemma to the crystallographic point groups and deduce that we have 32 trivial magnetic point groups (identical to the 32 crystallographic point groups), 32 paramagnetic groups, and $<3+3> + (5+13+5) + [5+13+8] + 3 = 58$ black-white point groups. This last sum is obtained by counting the number of lines connecting groups with subgroups of index 2 in Figure 3. The symbols $< >$, $( )$, $[ ]$ surrounding the figures refers to cubic, hexagonal-trigonal and tetragonal-orthorhombic groups; the absence of parenthesis for the last figure refers to monoclinic-triclinic point groups.

An important concept is that of "*admissible magnetic point groups*". It is clear that in magnetically ordered system, the magnetic point group of a magnetic atom cannot be one of the paramagnetic groups; moreover many of the colourless and black-white magnetic point groups cannot be realized in ordered system. Let us consider, for instance, the magnetic point group discussed above $\mathbf{M}_3 = 4'/m$. If we put a magnetic moment **S** in the origin pointing along an arbitrary direction and apply the operators, we arrive to the conclusion that **S** should vanish if $\mathbf{M}_3$ is to be a symmetry group. If **S** is along the fourfold axis, $4'$ inverts the spin so that $4'$ cannot be a symmetry operator. If **S** lies within the mirror plane, m inverts the spin so that m cannot be a symmetry operator. The point group $4'/m$ is an example of a *non-admissible* magnetic point group. There are only 31 admissible magnetic point groups that are listed, together with their admissible spin direction in the following table:

Admissible magnetic point groups					Admissible spin (magnetic moment) direction
1	$\bar{1}$				Any direction
2'	2'/m'	m'm2'			Perpendicular to the 2-fold axis (& to m for $m'm2'$)
m'					Any direction within the plane
m					Perpendicular to the plane
m'm'm					Perpendicular to the unprimed plane
2'2'2					Along the unprimed axis
2	2/m	m'm'2			Along the 2-fold axis
4	$\bar{4}$	4/m	42'2'		Along the four-fold axis
4m'm'	$\bar{4}2m'$		4/mm'm'		Along the four-fold axis
3	$\bar{3}$	32'	3m'	$\bar{3}m'$	Along the three-fold axis
6	$\bar{6}$	6/m	62'2'		Along the six-fold axis
6m'm'	$\bar{6}m'2'$		6/mm'm'		Along the six-fold axis

2.4: *Magnetic lattices*

A translation group in 3D is generated by three non-coplanar vectors $\{\mathbf{a}_1, \mathbf{a}_2, \mathbf{a}_3\}$ called the basis of the unit cell (sometimes we will use the crystallographic convention: $\{\mathbf{a}, \mathbf{b}, \mathbf{c}\}$). For primitive Bravais lattices, integer linear combinations of the three vectors generate the whole translation group: $\mathbf{T}_P = \{\mathbf{t} \mid \mathbf{t} = l_1\mathbf{a}_1 + l_2\mathbf{a}_2 + l_3\mathbf{a}_3, l_i \in \mathbb{Z}\}$

If we consider centred lattices the whole group is generated not only by the linear combinations but by adding centring vectors $\mathbf{t}_i^C (i = 1,..n_c)$:

$$\mathbf{T}_C = \{\mathbf{t} \mid \mathbf{t} = l_1\mathbf{a}_1 + l_2\mathbf{a}_2 + l_3\mathbf{a}_3 + n_1\mathbf{t}_1^C + n_2\mathbf{t}_2^C ... + n_{n_c}\mathbf{t}_{n_c}^C, l_i \in \mathbb{Z}, n_i \in \{0,1\}\}$$

The centring vectors are of the form: $\mathbf{t}_i^C = q_{1i}\mathbf{a}_1 + q_{2i}\mathbf{a}_2 + q_{3i}\mathbf{a}_3, (q_{ni} \in \mathbb{Q})$

The translation group can always be described using a primitive cell but, in order to simplify the expressions of the symmetry operators a centred basis is often more convenient. Following the previous procedure one can construct the different magnetic Bravais lattices. A subgroup, of index 2, of the translation group, can be obtained just by suppressing half of the translations; for instance if we take $l_1 = 2n$ we obtain a lattice $\mathbf{L}$ that is described with a unit cell with $\mathbf{a}_1' = 2\mathbf{a}_1$. This lattice is a subgroup of index 2 of $\mathbf{T}$ ($\mathbf{L} \subset \mathbf{T}$, $[i] = n(\mathbf{T})/n(\mathbf{L}) = 2$), the lost translations ($\mathbf{T}-\mathbf{L}$) can be associated with time reversal for constructing the magnetic lattice: $\mathbf{M_L} = \mathbf{L} + (\mathbf{T}-\mathbf{L})1'$. The classification of all magnetic lattices can be found in references [2] and [15].

3. Shubnikov Groups: Opechowski-Guccione and Belov-Neronova-Smirnova notations.

The same procedure can be applied to Bravais lattices and space groups. One obtains a total of 1651 types of Shubnikov groups. Among the 1651 magnetic space groups, considering $\mathbf{G}$ as a space group type, 230 are of the form $\mathbf{M_0}=\mathbf{G}$ (called also "monochrome"), 230 of the form $\mathbf{P}=\mathbf{G}+\mathbf{G}1'$ (paramagnetic or "grey" groups) and 1191 of the form $\mathbf{M}= \mathbf{H} + (\mathbf{G} - \mathbf{H})1'$ ("black-white" groups). Among the black-white groups there are 674 in which the subgroup $\mathbf{H} \subset \mathbf{G}$ is an equi-translation group: $\mathbf{H}$ has the same translation group as $\mathbf{G}$ (first kind, BW1). The rest of black-white groups, 517, are equi-class group (second kind, BW2). In this last family the translation subgroup contains "anti-translations" (pure translations associated with the spin reversal operator). The two notations for describing magnetic space groups existing in the literature are due to Belov-Neronova-Smirnova (BNS) [1] and to Opechowski-Guccione (OG) [2]. Both notations are identical for the major part of magnetic space groups except for the second kind black-white magnetic space groups. Recently a list of all magnetic space groups, in a similar form as that of the ITA for crystallographic groups, has been published [4] using the OG notation. A re-interpretation of [4] in terms of the BNS notation has also been published [5].

In the Shubnikov groups of the first kind the subgroup of translations is the same as that of the space group of which they derive, so the spin reversal operator is not associated with translations: the magnetic unit cell is the same as the crystallographic cell.
In the Shubnikov groups of the second kind there are some translations associated with spin reversal, so that the "primitive magnetic unit cell" is bigger than the primitive crystal cell.
The matrix representation of the symmetry operators depends on the basis used for referring to atom positions and magnetic moments. A drawback of using the magnetic unit cell as reference is that we have to re-write the symmetry operators and atom positions in a different basis as that of the crystallographic structure.

We shall use the ITA for determining all the magnetic groups deriving from a particular space group type. To be specific we select, as an example, the space group 46 *Ima*2. For constructing all magnetic space group derived from this space group we have to look for space groups that are subgroups of index 2. The symmetry operators of *Ima*2 and the subgroups are tabulated in ITA as follows:

Site symmetry and positions of space group *Ima*2

			(0, 0, 0)+	(½, ½, ½)+		
8	*c*	1	(1) x, y, z	(2) $-x, -y, z$	(3) $x+½, -y, z$	(4) $-x+½, y, z$
4	*b*	m . .	¼, y, z	¼, $-y, z$		
4	*a*	. . 2	0, 0, z	½, 0, z		

Symmetry operations

For (0, 0, 0)+ set
    (1) 1   (2) 2 0,0,z (3) a $x,0,z$ (4) m ¼, y,z
For (½ , ½ , ½)+ set
    (1) t(½ , ½ , ½)   (2) 2(0,0, ½)   ¼,¼,z (3) c x, ¼,z (4) n(0, ½ , ½) 0, y,z

       Maximal non-isomorphic subgroups of *Ima*2
     I   [2] *I* 1 1 2 (*C* 2) (1; 2)+
           [2] *I* 1 *a* 1   (*C c*) (1; 3)+
           [2] *I m* 1 1 (*C m*) (1; 4)+

     IIa   [2] *P m a* 2   1; 2; 3; 4
           [2] *P n c* 2   1; 2; (3; 4) + (½ , ½ , ½)
           [2] *P n a* 2$_1$ 1; 3; (2; 4) + (½ , ½ , ½)
           [2] *P m c* 2$_1$ 1; 4; (2; 3) + (½ , ½ , ½)
    IIb *none*

The groups I correspond to the *translationengleiche* of *t*-subgroups and the groups II to the *klassengleiche* or *k*-subgroups that are also subdivided in order to distinguish those having the same conventional cell (IIa) from those having a multiple cell (IIb) (see chapter from Tasci *et al.*). The division I and II correspond to the BW1 and BW2 respectively. Applying the formula **M** = **H** + (**G** − **H**) 1′ we obtain:

Colourless trivial magnetic group: **M**$_0$ = *I m a* 2 = {1, 2$_z$, a_y, m_x}**T**
For simplicity we remove the translation group and use only the coset representatives. We first list the BW1 groups using the data I of the ITA and show the OG notation that in this case coincides with the BNS notation. The subscript of mirror planes (rotation axes) indicates that they are perpendicular (parallel) to the given direction:

I :   **M**$_1$ = *I* 1 1 2 +(I m a 2 − *I* 1 1 2)1′ = {1, 2$_z$}+{ a_y, m_x}1′= *I m′ a′* 2
        M$_2$ = *I* 1 *a* 1 +(I m a 2 − *I* 1 *a* 1)1′ = {1, a_y}+{ 2$_z$, m_x}1′= *I m′ a* 2′
        M$_3$ = *I m* 1 1 +(I m a 2 − *I m* 1 1)1′ = {1, m_x }+{2$_z$, a_y}1′= *I m a′* 2′

For the BW2 groups, the translation subgroup is that formed by integer linear combinations of conventional cell parameters. The centring translations **t**$_C$ = (½, ½, ½) become anti-

translations (translations followed by a spin reversal: primed translations), so the magnetic lattice contains the following set of translations and anti-translations

$$\mathbf{L}= \{\mathbf{t} \mid \mathbf{t}=l_1\,\mathbf{a}_1 + l_2\,\mathbf{a}_2 + l_3\,\mathbf{a}_3;\ l_i \in \mathbb{Z}\}$$

$$\mathbf{L}'= (\mathbf{T}\text{-}\mathbf{L})1'= \{\mathbf{t}' \mid \mathbf{t}'=l_1\,\mathbf{a}_1 + l_2\,\mathbf{a}_2 + l_3\,\mathbf{a}_3 + \mathbf{t}_C;\ l_i \in \mathbb{Z}\}$$

We write below the BW2 groups using the data IIa of the ITA and show the BNS and the OG notation. It is clearly seen that the BNS notation correspond directly to the subgroups written in ITA by putting the lattice symbol P_I for stating that an original body centred lattice becomes primitive and the centring translations become anti-translations. None of the generators appearing in the symbol is primed. This is a characteristic of the BNS notation for all BW2 groups. On the contrary the OG conserves the original HM symbol changing the lattice type to I_P and using primed generators when they appear. From the information given in the decomposition of the magnetic group in terms of translations and anti-translations one can derive easily the BNS (the generators are selected from the coset representatives with respect to $\mathbf{L}$) or the OG notation (the generators are always those of the parent space group and they are primed if they belong to the coset representatives with respect to $\mathbf{L}'$).

IIa:

ITA	Notations:	BNS	OG
[2] *P m a* 2	$\mathbf{M}_4 = \{1, 2_z, a_y, m_x\}\mathbf{L} + \{1, 2_{1z}, c_y, n_x\}\mathbf{L}'$	$= P_I\,m\,a\,2$	$I_P\,m\,a\,2$
[2] *P n c* 2	$\mathbf{M}_5 = \{1, 2_z, c_y,\ n_x\}\mathbf{L} + \{1, 2_{1z}, a_y, m_x\}\mathbf{L}'$	$= P_I\,n\,c\,2$	$I_P\,m'\,a'\,2$
[2] *P n a* 2_1	$\mathbf{M}_6 = \{1, a_y, 2_{1z}, n_x\}\mathbf{L} + \{1, 2_z, c_y, m_x\}\mathbf{L}'$	$= P_I\,n\,a\,2_1$	$I_P\,m'\,a\,2'$
[2] *P m c* 2_1	$\mathbf{M}_7 = \{1, m_x, 2_{1z}, c_y\}\mathbf{L} + \{1, 2_z, a_y, n_x\}\mathbf{L}'$	$= P_I\,m\,c\,2_1$	$I_P\,m\,a'\,2'$

The association of spin reversal with translations is more conveniently described using the more general formalism of propagation vectors. Using this formalism we do not need the magnetic unit cell for describing the magnetic structure. Using the transformation properties of the spin configuration limited to the content of a primitive cell is enough to calculate the magnetic moment of whatever atom in the crystal.

4. Summary of the representation theory for space groups

For a mathematical treatment of the notions related to group theory and representation theory see chapter by Canals and Schober and that by Ballou. The group representation theory can be used to classify the possible magnetic structures that derive from a crystal structure in which there are magnetically ordered atoms. The magnetic order takes place in a crystal having symmetry properties described by a space group *G*.

Let us summarize the principal results of the representation group theory. If a mapping can be established between the elements of a group and a set of square matrices that preserves the group multiplication, we say that the set of matrices constitute a "representation" of the group.

$$\Gamma = \Gamma(g) \mid g \in G\ ,\quad \Gamma(g_1 g_2) = \Gamma(g_1)\Gamma(g_2) \tag{6}$$

A similarity transformation applied to all matrices provides an equivalent representation (the matrix *U* is generally unitary: $U^{-1}=U^{\dagger}$).

$$\Gamma(g) = U\,\Gamma(g)\,U^{-1} \quad \text{with } g \in G \tag{7}$$

A particular group has an infinite number of representations of arbitrary dimensions. An arbitrary representation may be reduced to "block-diagonal form" by an appropriate similarity transformation. For instance the shape of all the matrices of the representation after the transformation may be as:

$$\Gamma(g) = \begin{pmatrix} A_{11} & A_{12} & 0 & 0 & 0 & 0 & 0 \\ A_{21} & A_{22} & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & B_{11} & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & B_{11} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & C_{11} & C_{12} & C_{13} \\ 0 & 0 & 0 & 0 & C_{21} & C_{22} & C_{23} \\ 0 & 0 & 0 & 0 & C_{31} & C_{32} & C_{33} \end{pmatrix} = A(g) \oplus 2B(g) \oplus C(g) \tag{8}$$

The matrices A(g), B(g) and C(g) are also representations of the group. The most important representations are called "Irreducible Representations" (*irreps*). These are those that cannot be reduced to block-diagonal form. A reducible representation can be decomposed in direct sum of *irreps* in a unique way. We shall note the different *irreps* with the index ν and a symbol Γ that may be used also for matrices. The decomposition of a reducible representation can be written as:

$$\Gamma = \sum_{\oplus \nu} n_\nu \Gamma^\nu = n_1 \Gamma^1 \oplus n_2 \Gamma^2 \oplus n_3 \Gamma^3 ... \oplus n_m \Gamma^m \tag{9}$$

The dimension of the representation Γ^ν is l_ν. The characters of a representation (traces of the matrices) will be represented as $\chi_\nu(g)$. The irreducible representations satisfy the following great orthogonality theorem:

$$\sum_{g \in G} \Gamma_{ij}^{\nu*}(g) \Gamma_{lm}^{\mu}(g) = \frac{n(G)}{l_\nu} \delta_{il}\,\delta_{jm}\,\delta_{\mu\nu} \tag{10}$$

with $n(G)$ the order of the group. We can deduce the following corollary for the characters of the *irreps*:

$$\sum_{g \in G} \chi^{\nu*}(g)\chi^{\mu}(g) = n(G)\,\delta_{\mu\nu} \tag{11}$$

As a consequence we can calculate the number of times a particular *irrep* is contained in a reducible representation. From (9) and (11):

$$n_\nu = \frac{1}{n(G)} \sum_{g \in G} \chi(g)\chi^{*\nu}(g) \tag{12}$$

The elements of the symmetry groups act on position vectors. For each particular problem we can select a set of physically relevant variables $\varphi_i \{i = 1, 2 \ldots p\}$ spanning a working functional space **W**. These functions constitute a basis of the **W** space. The action of the operator associated to a symmetry operator when applied to a function of position vectors is defined by the expression:

$$O(g)\varphi(\mathbf{r}) = \varphi(g^{-1}\mathbf{r}) \equiv \varphi'(\mathbf{r}) \tag{13}$$

When using the functions $\varphi_i(\mathbf{r})$, the action of the operator $O(g)$ gives rise to a linear combination, defining a representation of the group **G**:

$$O(g)\varphi_j(\mathbf{r}) = \varphi'(\mathbf{r}) = \sum_i \Gamma_{ij}(g)\varphi_i(\mathbf{r}) \tag{14}$$

If we take another basis ψ related to the initial one by a unitary transformation we may get the matrices of the Γ representation in block-diagonal form.

$$\psi_j(\mathbf{r}) = \sum_i U_{ij}\varphi_i(\mathbf{r}) \tag{15}$$

The system of p ψ-functions splits in subsystems defining irreducible subspaces of the working space **W**. If we take one of these subspaces (labelled ν), the action of the operator $O(g)$ on the basis functions is:

$$O(g)\psi_j(\mathbf{r}) = \sum_{i=1}^{l_\nu} \Gamma_{ij}^\nu(g)\psi_i(\mathbf{r}) \tag{16}$$

Here the functions are restricted to those of the subspace ν.
In treating a physical problem for a symmetry analysis, it is simple to find the initial working space with an arbitrary set of basis functions that generates a reducible representation. We dispose of a set of operators, called *projection operators*, providing the basis functions of the irreducible representations contained in the initial reducible representation. The action of the projection operator P^ν providing the basis functions of the *irrep* Γ^ν is given by the expression:

$$\psi_i^\nu = P^\nu\varphi = \frac{1}{n(G)} \sum_{g \in G} \Gamma_{i[j]}^{*\nu}(g)\, O(g)\, \varphi \qquad (i = 1, \ldots l_\nu) \tag{17}$$

The result of the above operation is zero or a basis function of the corresponding *irrep*. The index [*j*] is fixed, taking different values it provides new basis functions or zero.

This is all we need for our purposes. We shall study how the so called magnetic representation decomposes in irreducible representations of space groups. For that let us start working with the translation subgroup of whatever space group. The translation group $\mathbf{T} \subset \mathbf{G}$ is Abelian (commutative), so their *irreps* are all *one-dimensional*. Considering the properties of the translation operators and the Born-Von Karman periodic boundary conditions (see chapter by Schober), the translation operators and their representation matrix (a single number equal to its character) are given by the expressions:

$$O(\mathbf{t}) = O(l_1\mathbf{a}_1 + l_2\mathbf{a}_2 + l_3\mathbf{a}_3) = O(\mathbf{a}_1)^{l_1} O(\mathbf{a}_2)^{l_2} O(\mathbf{a}_3)^{l_3}$$

$$O(\mathbf{a}_j)^{N_j+1} = O(\mathbf{a}_j) \qquad j = 1, 2, 3 \tag{18}$$

$$O(\mathbf{t}) \rightarrow \exp\left\{2\pi i\left(\frac{p_1 l_1}{N_1} + \frac{p_2 l_2}{N_2} + \frac{p_3 l_3}{N_3}\right)\right\}, \qquad p_i, N_i \in \mathbb{Z} \ \& \ 0 \le p_i \le N_i - 1$$

N_i is the number of unit cells along the direction $\mathbf{a}_i$. There are $N_1 \times N_2 \times N_3 = N$ representations (= number of unit cells in the crystal) labelled by the reciprocal space vector:

$$\mathbf{k} = \left(\frac{p_1}{N_1}, \frac{p_2}{N_2}, \frac{p_3}{N_3}\right) = \frac{p_1}{N_1}\mathbf{b}_1 + \frac{p_2}{N_2}\mathbf{b}_2 + \frac{p_3}{N_3}\mathbf{b}_3 \tag{19}$$

The matrix of the representation $\mathbf{k}$ corresponding to the translation $\mathbf{t}$ is then:

$$\Gamma^{\mathbf{k}}(\mathbf{t}) = \exp\left\{2\pi i\left(\frac{p_1 l_1}{N_1} + \frac{p_2 l_2}{N_2} + \frac{p_3 l_3}{N_3}\right)\right\} = \exp\ 2\pi i\mathbf{k}\,\mathbf{t} \tag{20}$$

where the $\mathbf{k}$-vectors in reciprocal space are restricted to the first Brillouin Zone[2].
It is clear that adding a reciprocal lattice vector $\mathbf{H}$ to $\mathbf{k}$ does not change the matrix, so the vectors $\mathbf{k'}=\mathbf{H}+\mathbf{k}$ and $\mathbf{k}$ are equivalent.
The basis functions of the group of translations must satisfy the equation:

$$O(\mathbf{t})\psi^{\mathbf{k}}(\mathbf{r}) = \Gamma^{\mathbf{k}}(\mathbf{t})\psi^{\mathbf{k}}(\mathbf{r}) = \exp\ 2\pi i\mathbf{k}\,\mathbf{t}\ \psi^{\mathbf{k}}(\mathbf{r}) \tag{21}$$

The most general form for the functions $\psi^{\mathbf{k}}(\mathbf{r})$ are the Bloch functions[3]:

$$\psi^{\mathbf{k}}(\mathbf{r}) = u_{\mathbf{k}}(\mathbf{r})\exp\{-2\pi i\mathbf{k}\mathbf{r}\}, with\ u_{\mathbf{k}}(\mathbf{r} \pm \mathbf{t}) = u_{\mathbf{k}}(\mathbf{r}) \tag{22}$$

For constructing the representations of the space groups it is important to start with the basis functions. Let us see how the Bloch functions behave under the action of a general element of the space group $g=\{h|\mathbf{t}_h\}$:

$$O(g)\psi^{\mathbf{k}}(\mathbf{r}) = \{h\,|\,\mathbf{t}_h\}\psi^{\mathbf{k}}(\mathbf{r}) = \psi\,'(\mathbf{r}) \tag{23}$$

To determine the form of the functions $\psi\,'(\mathbf{r})$ one can see that they should also be Bloch functions with a different $\mathbf{k}$-label:

$$O(\mathbf{t})\psi\,'(\mathbf{r}) = \{1\,|\,\mathbf{t}\}\psi\,'(\mathbf{r}) = \{1\,|\,\mathbf{t}\}\{h\,|\,\mathbf{t}_h\}\psi^{\mathbf{k}}(\mathbf{r}) = \{h\,|\,\mathbf{t}_h\}\{1\,|\,h^{-1}\mathbf{t}\}\psi^{\mathbf{k}}(\mathbf{r}) =$$
$$= \{h\,|\,\mathbf{t}_h\}\exp\{2\pi i\mathbf{k}\,h^{-1}\mathbf{t}\}\psi^{\mathbf{k}}(\mathbf{r}) = \exp\{2\pi i\mathbf{k}\,h^{-1}\mathbf{t}\}\{h\,|\,\mathbf{t}_h\}\psi^{\mathbf{k}}(\mathbf{r}) = \tag{24}$$
$$= \exp\{2\pi i\,h\mathbf{k}\,\mathbf{t}\}\psi\,'(\mathbf{r})$$

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[2] The sign of the phase factor is a matter of convention. Here we take the positive sign in order to be consistent with the convention used in crystallography for Fourier series. This selection makes the phase sign of Bloch functions to be negative (see chapter by Schober). In many textbooks of solid state physics and representation analysis the negative sign is adopted. This reduces to rename the vector $\mathbf{k}$ as $-\mathbf{k}$ and vice versa.
[3] This is easily verified by applying the rules of the operators action on functions:
$O(\mathbf{t})\psi^{\mathbf{k}}(\mathbf{r}) = \psi^{\mathbf{k}}(\mathbf{r}-\mathbf{t}) = u_{\mathbf{k}}(\mathbf{r}-\mathbf{t})\exp\{-2\pi i\mathbf{k}(\mathbf{r}-\mathbf{t})\} = \exp\{2\pi i\mathbf{k}\mathbf{t}\}u_{\mathbf{k}}(\mathbf{r})\exp\{-2\pi i\mathbf{k}\mathbf{r}\} = \exp\{2\pi i\mathbf{k}\mathbf{t}\}\psi^{\mathbf{k}}(\mathbf{r})$

so that:

$$O(g)\psi^{\mathbf{k}}(\mathbf{r}) = \{h \mid \mathbf{t}_h\}\psi^{\mathbf{k}}(\mathbf{r}) = \psi^{h\mathbf{k}}(\mathbf{r}) \qquad (25)$$

The Bloch functions also serve as basis functions but the representations are no longer one-dimensional because the Bloch functions whose wave vectors are related by the rotational part of $g \in \mathbf{G}$ belong to a same subspace. The set of non-equivalent $\mathbf{k}$ vectors obtained by applying the rotational part of the symmetry operators of the space group constitute the so called "star of $\mathbf{k}$":

$$\{\mathbf{k}\} = \{h_1\mathbf{k}_1, h_2\mathbf{k}_1, h_3\mathbf{k}_1, h_4\mathbf{k}_1, \ldots\} = \{\mathbf{k}_1, \mathbf{k}_2, \ldots \mathbf{k}_{l_k}\} \qquad (26)$$

The $\mathbf{k}_i$ vectors are called the arms of the star. The number l_k is less or equal to the order of the point group $n(G_0)$. The set of elements $g \in \mathbf{G}$ leaving the $\mathbf{k}$ vector invariant, or equal to an equivalent vector, form the group $\mathbf{G_k}$, called the group of the wave vector (or propagation vector group) or the "little group". It is always a subgroup of $\mathbf{G}$. The whole space/point group can be decomposed in cosets of the propagation vector group:

$$Space\,Group: \qquad \mathbf{G} = \mathbf{G_k} + g_2\mathbf{G_k} + \ldots = \sum_{L=1}^{l_k} g_L\mathbf{G_k}$$

$$Point\,Group: \qquad \mathbf{G}_0 = \mathbf{G_{0k}} + h_2\mathbf{G_{0k}} + \ldots = \sum_{L=1}^{l_k} h_L\mathbf{G_{0k}} \qquad \mathbf{k}_L = h_L\mathbf{k} \qquad (27)$$

The point group $\mathbf{G_{0k}}$ is also called the "little co-group". The fact that the translation group is a normal subgroup of the space group simplifies enormously the representations of space groups. We have to handle only the representative elements of the space group in their decomposition into cosets of the translation group. The quotient group $\mathbf{G}/\mathbf{T}$ is always finite and isomorphic to the point group of the space group.

Let us note the irreducible representations of $\mathbf{G_k}$ as $\Gamma^{\mathbf{k}\nu}$ of dimensionality l_ν. The basis functions should be of the form:

$$\psi_i^{\mathbf{k}\nu}(\mathbf{r}) = u_{\mathbf{k}i}^{\nu}(\mathbf{r})\exp\,-2\pi i\,\mathbf{k}\,\mathbf{t} \qquad (i = 1, 2 \ldots l_\nu) \qquad (28)$$

Under the action of the elements of $\mathbf{G_k}$ the functions transform into each other with the same $\mathbf{k}$-vector. Using the elements of $\mathbf{G}$ not belonging to $\mathbf{G_k}$ one generates other sets of basis functions:

$$\psi_i^{\mathbf{k}_1\nu}(\mathbf{r}); \psi_i^{\mathbf{k}_2\nu}(\mathbf{r}); \psi_i^{\mathbf{k}_3\nu}(\mathbf{r}) \quad \ldots \quad \psi_i^{\mathbf{k}_L\nu}(\mathbf{r}); \qquad (i = 1, 2 \ldots l_\nu) \qquad (29)$$

constituting the basis functions of the representations of the total space group.
These representations are labelled by the star of the $\mathbf{k}$ vector as: $\Gamma^{\{\mathbf{k}\}\nu}$ and are of dimensionality $l_\nu \times l_k$. Each irreducible "small representation" induces an irreducible representation of the total space group. The *induction formula* is:

$$\Gamma_{Li,Mj}^{\{\mathbf{k}\}\nu}(g) = \Gamma_{ij}^{\mathbf{k}\nu}(g_L^{-1}g\,g_M)\delta_{g_L^{-1}g\,g_M \in G_{\mathbf{k}}} \qquad (30)$$

The symbol δ is 1 if the subscript condition is true, otherwise it is zero.

We need to know only the matrices of the small representations (representations of $\mathbf{G_k}$) for the coset representatives of $\mathbf{G_k}$ with respect to the translation group:

$$\mathbf{G_k} = 1\mathbf{T} + g_2\mathbf{T} + g_3\mathbf{T} + ... + g_n\mathbf{T} \tag{31}$$

For a general element of $\mathbf{G_k}$ we have:

$$\Gamma^{\mathbf{k}\nu}(g) = \Gamma^{\mathbf{k}\nu}(\{h\,|\,\mathbf{t}_h + \mathbf{t}\}) = \Gamma^{\mathbf{k}\nu}(\{1\,|\,\mathbf{t}\}\{h\,|\,\mathbf{t}_h\}) = \Gamma^{\mathbf{k}\nu}(\{1\,|\,\mathbf{t}\})\ \Gamma^{\mathbf{k}\nu}(\{h\,|\,\mathbf{t}_h\})$$
$$\Gamma^{\mathbf{k}\nu}(\{h\,|\,\mathbf{t}_h + \mathbf{t}\}) = e^{2\pi i\mathbf{k}\mathbf{t}}\ \Gamma^{\mathbf{k}\nu}(\{h\,|\,\mathbf{t}_h\}) \tag{32}$$

The matrices $\Gamma^{\mathbf{k}\nu}$ of the small representations can be easily calculated from the projective (or *loaded*) representations that are tabulated in the Kovalev book [16]:

$$\Gamma^{\mathbf{k}\nu}(g) = \Gamma^{\mathbf{k}\nu}(\{h\,|\,\mathbf{t}_h\}) = \Gamma^{\nu}_{proj}(h)\,e^{2\pi i\mathbf{k}\mathbf{t}_h} \tag{33}$$

or calculated with the appropriate algorithm [17].

5. The formalism of propagation vectors for describing magnetic structures.

If we disregard, for the moment, the symmetry properties, except the translation subgroup of the space group, of the magnetic moment configuration whatever class of magnetic structure can be represented by the Fourier series:

$$\mathbf{m}_{lj} = \sum_{\mathbf{k}} \mathbf{S}_{\mathbf{k}j}\,exp(-2\pi i\mathbf{k}\mathbf{R}_l) \tag{34}$$

This defines the magnetic moment of the atom numbered j in the unit cell having as origin the lattice vector $\mathbf{R}_l$ (the atom at $\mathbf{R}_{lj} = \mathbf{R}_l + \mathbf{r}_j$). The $\mathbf{k}$ vectors are defined in reciprocal space and are called propagation vectors of the magnetic structure. For the description of magnetic structures they can be restricted to the first Brillouin zone (BZ). Notice that a Fourier series in nothing else that a development of a function in terms of Bloch functions.
The Fourier coefficients $\mathbf{S}_{\mathbf{k}j}$ are, in general, complex vectors and must verify the equality $\mathbf{S}_{\mathbf{k}j} = \mathbf{S}^*_{-\mathbf{k}j}$ to make the sum result a real vector. Even disordered magnetic structures, like those of *spin glasses*, may eventually be described with an expression like (34) if a nearly continuous distribution of $\mathbf{k}$-vectors inside the BZ is considered. In practice, most of the magnetic structures can be describe by a small number (1 to 3) of propagation vectors.

We may have defined (34) in a slightly different manner also usual in literature. Instead of writing $\mathbf{R}_l$ in the argument of the exponential function, one can write $\mathbf{R}_{lj} = \mathbf{R}_l + \mathbf{r}_j$:

$$\mathbf{m}_{lj} = \sum_{\mathbf{k}} \mathbf{T}_{\mathbf{k}j}\,exp(-2\pi i\mathbf{k}(\mathbf{R}_l + \mathbf{r}_j)) \tag{34'}$$

In such a case the Fourier coefficients, $\mathbf{T}_{\mathbf{k}j}$, of the new expression should be related to those of expression (34) by a phase factor, $\mathbf{S}_{\mathbf{k}j} = \mathbf{T}_{\mathbf{k}j}\,exp(-2\pi i\mathbf{k}\mathbf{r}_j)$, that depends on the atom positions inside the unit cell. We shall see that the convention we have adopted (34), similar to that of reference [18], is more convenient for a unified description of commensurate and incommensurate magnetic structures. On the other hand (34') is more convenient when symmetry invariance is used in incommensurate structures.

The formulae we are considering in this paragraph are written in vectorial form, so they are independent of the particular frame for describing the magnetic moments, propagation vectors, atom positions, etc. Atom positions inside the unit cell are normally referred to the conventional basis $A=(\mathbf{a}, \mathbf{b}, \mathbf{c})$, so that fractional co-ordinates are dimensionless. The Fourier coefficients, $\mathbf{S}_{\mathbf{k}j}$, have the same units as magnetic moments, usually Bohr magnetons, and their components are given with respect to the unitary frame of the conventional unit cell $U=(\mathbf{a}/a, \mathbf{b}/b, \mathbf{c}/c) = (\mathbf{e}_1, \mathbf{e}_2, \mathbf{e}_3)$. The components of reciprocal lattice vectors, $\mathbf{H}$, of the nuclear structure and those of the propagation vectors $\mathbf{k}$, are given with respect to the reciprocal basis of the conventional unit cell and are also dimensionless. This is a very important point because in many of the available tables of space group irreducible representations, the $\mathbf{k}$-vectors are given with respect to a *primitive basis* of the reciprocal lattice $b=(\mathbf{b}_1, \mathbf{b}_2, \mathbf{b}_3)$ which coincides with $a^*=(\mathbf{a}_1{}^*, \mathbf{a}_2{}^*, \mathbf{a}_3{}^*)$ only for primitive direct lattices. When the Bravais lattice of the crystal is centred, the set b is obtained from a primitive basis of the direct cell and not from the Bravais (or conventional) unit cell. Izyumov and collaborators [15] have introduced a set of vectors $B=(\mathbf{B}_1, \mathbf{B}_2, \mathbf{B}_3)$ as a reference frame for the reciprocal lattice. The set B correspond to the "Bravais cell" of the reciprocal lattice. This is a frame that is not used by crystallographers and, giving the components of the reciprocal vectors with respect to it is confusing. We do not recommend the use of this frame for studies of magnetic structures.

The lattice vectors $\mathbf{R}_l$ appearing in the argument of the exponential function are integer linear combinations of the basis vectors of frame A only for primitive lattices. For centred lattices two formal types of lattice vectors exist: $\mathbf{R}_l = \mathbf{R}_n = n_1\mathbf{a} + n_2\mathbf{b} + n_3\mathbf{c}$ with $n_i \in \mathbb{Z}$ and $\mathbf{R}_l = \mathbf{R}_n + \mathbf{t}_C$, where $\mathbf{t}_C$ is one of the centring vectors of the lattice with components $t_i \in \mathbb{Q}$. There is nothing special for these two types of lattice vectors, the different type of components is just a matter of convention. The minimum set of magnetic atoms to be considered for describing a magnetic structure, without considering at this stage the symmetry, should not be related by lattice centring translations. The expression (34) assures the knowledge of the magnetic moments in the whole crystal when we know both the propagation vector(s) and the Fourier coefficients of the elemental set of atoms in the reference zero-cell $\mathbf{R}_0 = (0,0,0)$. The translational properties of the crystal allow reducing the necessary number of free parameters for describing a magnetic structure. If no translational properties exist the free parameters correspond to the three components of magnetic moments of each atom in the crystal. The Fourier decomposition (34), consequence of the translational symmetry, indicates that we need to consider only a finite number of, in general, complex vectors associated to the magnetic atoms within a primitive unit cell.

Let us describe general types of magnetic structures of increasing degree of complexity, using the formalism of propagation vectors through the expression (34).

a) The simplest types of magnetic structures existing in complex crystals have a single null propagation vector at the centre of the BZ: $\mathbf{k} = (0, 0, 0) = \mathbf{0}$. The Fourier coefficients should be real and can be identified to the magnetic moments directly:
$$\mathbf{m}_{lj} = \mathbf{S}_{\mathbf{0}j}exp(-2\pi i \mathbf{0} \mathbf{R}_l) = \mathbf{S}_{\mathbf{0}j} = \mathbf{m}_{\mathbf{0}j} \qquad (35)$$
This expression tells us that the orientation and magnitudes of the magnetic moments in whatever cell of the crystal are identical to those of the zero-cell. The translational symmetry of the magnetic structure is identical to that of the crystal structure: the magnetic unit cell is the same as the chemical cell. This class of magnetic structures may be ferromagnetic, ferrimagnetic or antiferromagnetic, collinear or non-collinear. The propagation vector at the

centre of the BZ does not mean that the magnetic structure is ferromagnetic. This is only true for Bravais lattices (a single magnetic atom per primitive cell). This kind of magnetic structure can be described by one of the 230 monochrome or the 674 black-white first kind (BW1) magnetic space groups.

Notice that if we had taken the convention for the Fourier series (34) that puts in the exponential term the global vector position of the atom, $\mathbf{R}_{lj} = \mathbf{R}_l + \mathbf{r}_j$, the Fourier coefficients $\mathbf{T}_{\mathbf{k}j}$ could not be identified with magnetic moments because of the phase factor containing the atom positions.

b) The next class of magnetic structures corresponds also to a single propagation vector, in this case of the form: $\mathbf{k}=1/2\mathbf{H}$, where $\mathbf{H}$ is a reciprocal lattice vector. The propagation vectors of this kind correspond to high symmetry points of the surface of the BZ (Liftchitz points). In this case we have:

$$\mathbf{m}_{lj} = \mathbf{S}_{\mathbf{k}j} exp(-2\pi i \mathbf{k} \mathbf{R}_l) = \mathbf{S}_{\mathbf{k}j} exp(-\pi i \mathbf{H} \mathbf{R}_l) = \mathbf{S}_{\mathbf{k}j}(-1)^{\mathbf{H}\mathbf{R}_l} = \mathbf{S}_{\mathbf{k}j}(-1)^{n_l} = \mathbf{m}_{0j}(-1)^{n_l} \tag{36}$$

This expression tells us that the orientation and magnitudes of the magnetic moments in whatever cell of the crystal are either identical or opposite to those of the zero-cell. The translational symmetry is lower than that of the chemical cell. The magnetic cell can easily be deduced from the particular values of the propagation vector (see [15] for a classification of magnetic lattices in terms of propagation vectors). The magnetic structures of this kind are necessarily antiferromagnetic and can be described by one of the 517 black-white second kind (BW2) magnetic space groups.

c) This is the general case, where the $\mathbf{k}$-vector is not a special vector as in the two previous types. For these cases there is no Shubnikov group, in three dimensions, that can describe the symmetry properties of such spin configuration. The general expression of the Fourier coefficient for the atom j is explicitly given by:

$$\mathbf{S}_{\mathbf{k}j} = \frac{1}{2}\{\vec{R}_{\mathbf{k}j} + i\vec{I}_{\mathbf{k}j}\}\exp(-2\pi i\phi_{\mathbf{k}j}) = \frac{1}{2}\{R_{\mathbf{k}j}^x\mathbf{e}_1 + R_{\mathbf{k}j}^y\mathbf{e}_2 + R_{\mathbf{k}j}^z\mathbf{e}_3 + i(I_{\mathbf{k}j}^x\mathbf{e}_1 + I_{\mathbf{k}j}^y\mathbf{e}_2 + I_{\mathbf{k}j}^z\mathbf{e}_3)\}\exp(-2\pi i\phi_{\mathbf{k}j})$$

Only six real parameters define the $\mathbf{S}_{\mathbf{k}j}$ vectors, so the phase factor $\phi_{\mathbf{k}j}$ is not generally needed, but it is convenient to use it when particular relations or constraints between real and imaginary vectors, $(\vec{R}_{\mathbf{k}j}, \vec{I}_{\mathbf{k}j})$, are given. The calculation of the magnetic moment of the atom j in the unit cell of index l, should be performed by using the formula (34) that may be also written in this case as:

$$\mathbf{m}_{lj} = \sum_{\langle\mathbf{k}\rangle}\{\vec{R}_{\mathbf{k}j}\cos 2\pi[\mathbf{k}\mathbf{R}_l + \phi_{\mathbf{k}j}] + \vec{I}_{\mathbf{k}j}\sin 2\pi[\mathbf{k}\mathbf{R}_l + \phi_{\mathbf{k}j}]\} \tag{37}$$

where the sum is now extended to half the number of propagation vectors, i.e. over the total number of pairs ($\mathbf{k},-\mathbf{k}$).

If the magnetic structure represents a *helical* order the Fourier coefficients are of the form:

$$\mathbf{S}_{\mathbf{k}j} = \frac{1}{2}\{m_{1j}\mathbf{u}_j + i m_{2j}\mathbf{v}_j\}\exp(-2\pi i\phi_{\mathbf{k}j}), \quad with \quad |\mathbf{u}_j|=|\mathbf{v}_j|=1, \quad \mathbf{u}_j\cdot\mathbf{v}_j=0 \tag{38}$$

where $\mathbf{u}_j$ and $\mathbf{v}_j$ are orthogonal unit vectors.

Three situations can then be encountered:

i- $m_{2j} = 0$: The magnetic structure corresponds to a modulated sinusoid of amplitude $A = m_{1j}$

ii- $m_{1j} = m_{2j}$: All *j* atoms have equal magnetic moments. If the propagation vector is perpendicular to the plane formed by the vectors $\mathbf{u}_j$ and $\mathbf{v}_j$ the magnetic structure for the sublattice *j* corresponds to a classical *helix* (or *spiral*), of cylindrical envelope (see Fig. 4b). If the propagation vector is within the (**u**, **v**) plane, the structure is called a *cycloid* (see Fig. 4c).

iii- $m_{1j} \neq m_{2j}$: The *helix* (or *cycloid*) has an elliptical envelope and the moments have values between min(m_{1j}, m_{2j}) and max(m_{1j}, m_{2j}).

Figure 4: Examples of magnetic structures. In all cases the orientation of the lattice is similar, except in the second view of (b) in which the point of view is nearly along **b**.
  (a) Sinusoidal structure with propagation vector **k**=(0,δ,0) and $\mathbf{S_k}$=(0,0,w)
  (b) Helical, or spiral, structure with propagation vector **k**=(0,δ,0) and $\mathbf{S_k}$=(u*i*,0,u)
  (c) Cycloidal structure with propagation vector **k**=(0,δ,0) and $\mathbf{S_k}$=(0,u,u*i*)
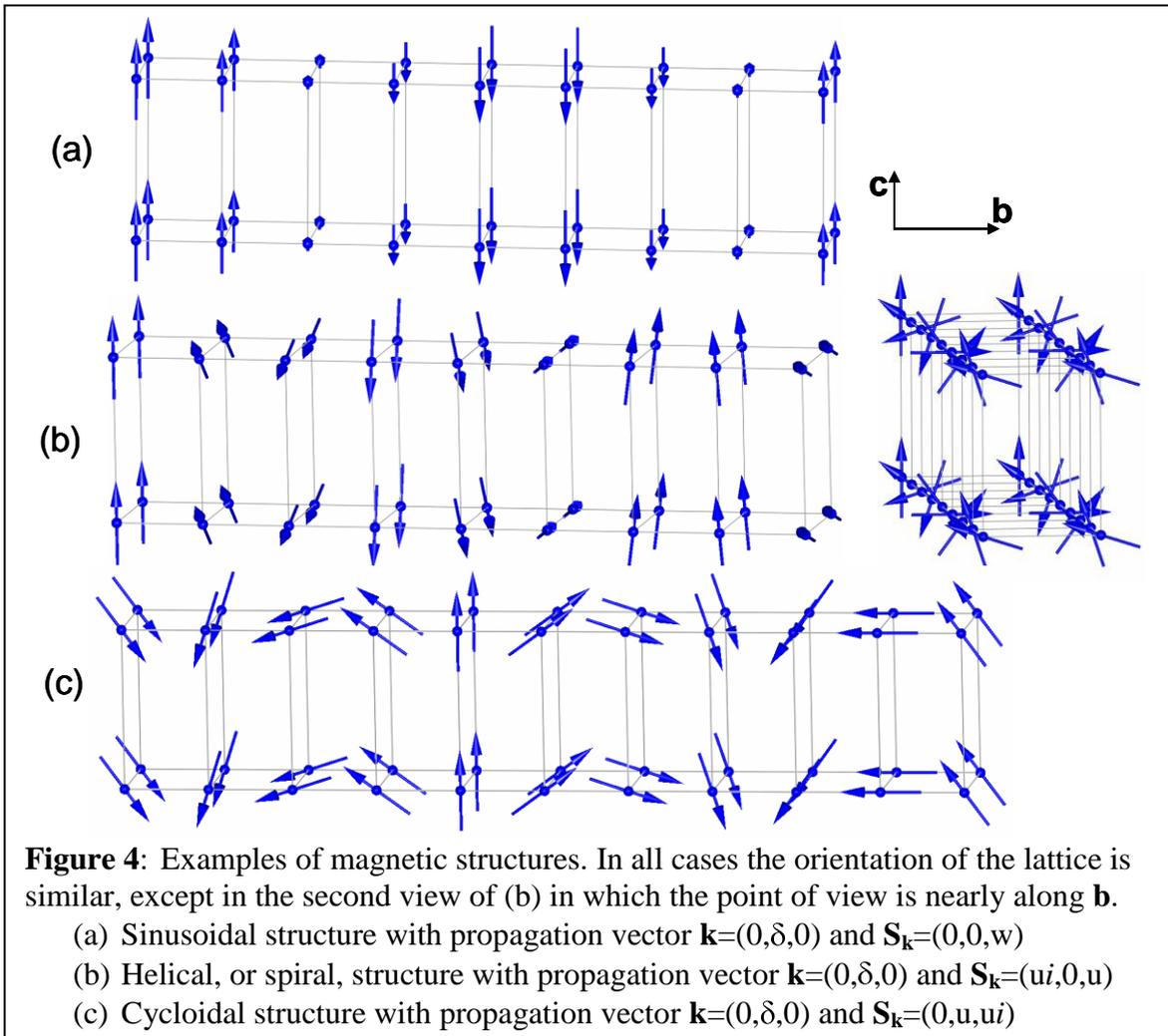
6. How we get information about magnetic structures: magnetic neutron scattering

6.1: *The scattering amplitude*

The intensity of magnetic Bragg peaks due to neutron scattering by magnetically ordered systems can be calculated in a similar way as for X-rays or nuclear neutron scattering. The most important difference is that the scattering amplitude is not a scalar variable. Here we will give a summary of the most important expressions needed to calculate the intensity of a Bragg

reflection, the reader interested in more details of the elastic magnetic scattering in relation with magnetic structures can consult [18] and the references therein.

The interaction of neutrons with the magnetic moments of atoms is of dipolar origin through the magnetic moment of the neutron. The scattering amplitude vector, for a single atom with atomic moment **m**, is given by:

$$\mathbf{a}(\mathbf{Q}) = pf(Q)\mathbf{m}_\perp = \frac{1}{2}r_e\gamma f(Q)\left(\mathbf{m} - \frac{\mathbf{Q}(\mathbf{m}\cdot\mathbf{Q})}{Q^2}\right) = \frac{1}{2Q^2}r_e\gamma f(Q)(\mathbf{Q}\times\mathbf{m}\times\mathbf{Q}) \qquad (39)$$

The constant $p = r_e\ \gamma/2 = 0.2695$ allows the conversion of the magnetic moments, given in Bohr magnetons to scattering lengths units of 10^{-12} cm. The other constants appearing in formula (39) are: the classical radius of the electron $r_e = e^2/mc^2 = 2.81776\times10^{-13}$ cm and the magnetic moment of the neutron in nuclear magnetons γ (=1.9132). The function $f(Q)$ is the atomic magnetic form-factor (Fourier transform of the unpaired electron density, normalized as $f(0) = 1$, assumed to be spherical hereafter), and $\mathbf{m}_\perp$ is the perpendicular component of the atomic moment to the scattering vector $\mathbf{Q}=2\pi\mathbf{s}$. As shown in (39), only the perpendicular component of **m** contributes to the magnetic scattering of neutrons by matter. The vectorial character of the interaction allows determining the magnetic moment direction with respect to the crystal lattice.

6.2: *Magnetic interaction vector of the whole crystal*

For unpolarised neutrons the nuclear and magnetic intensities are simply additive and are, in general, of the same order of magnitude. An important difference between nuclear and magnetic scattering is that the latter is strongly reduced at high Q. The absence of "form factor" in the case of nuclear scattering gives rise to a decrease of intensity with Q due only to thermal vibrations, whereas the magnetic form factor $f(Q)$ in (39) is the Fourier transform of the unpaired electron density having a spatial extension of the order of magnitude of the neutron wavelength. The elastic scattered intensity by a crystal, as a function of **Q** or **s**, is proportional to the square of the total amplitude (also called "magnetic interaction vector"):

$$\mathbf{M}_\perp^T(\mathbf{s}) = \sum_{lj} pf_j(s)\mathbf{m}_{\perp lj}e^{2\pi i\mathbf{s}\cdot\mathbf{R}_{lj}} = \frac{p}{s^2}\mathbf{s}\times\sum_{lj}f_j(s)\mathbf{m}_{lj}e^{2\pi i\mathbf{s}\cdot\mathbf{R}_{lj}}\times\mathbf{s} = \frac{1}{s^2}\mathbf{s}\times\mathbf{M}^T(\mathbf{s})\times\mathbf{s} \quad (40)$$

The vector $\mathbf{M}^T$ is the magnetic structure factor of the whole crystal. The scattered intensity is calculated multiplying the expression (40) by its complex conjugate as usual.

Contrary to the majority of crystal structures, many magnetic structures are incommensurate: the periodicity of the orientation of the magnetic moments is not commensurate with the underlying crystal structure. This is a consequence of the competition of the exchange interactions giving rise to different kinds of frustration in many compounds. We shall develop the expression (40) using the formalism of propagation vectors that allows, for magnetic scattering, a general way of treating, on the same foot, commensurate and incommensurate magnetic structures. Using this formalism we do not need to use the concept of magnetic unit cell even in the case of commensurate structures.

6.3: *The magnetic structure factor*

We can substitute (34) in the expression (40) to obtain a compact description of the magnetic interaction vector of the whole crystal:

$$\mathbf{M}^T(\mathbf{s}) = p\sum_{lj} f_j(s)\mathbf{m}_{lj}e^{2\pi i\mathbf{s}\cdot\mathbf{R}_{lj}} = p\sum_{lj} f_j(s)e^{2\pi i\mathbf{s}\cdot\mathbf{R}_{lj}}\sum_{\mathbf{k}}\mathbf{S}_{\mathbf{k}j}e^{-2\pi i\mathbf{k}\cdot\mathbf{R}_l} =$$

$$= p\sum_{j} f_j(s)e^{2\pi i\mathbf{s}\cdot\mathbf{r}_j}\sum_{\mathbf{k}}\mathbf{S}_{\mathbf{k}j}\sum_{l}e^{2\pi i(\mathbf{s}-\mathbf{k})\cdot\mathbf{R}_l} \propto p\sum_{j} f_j(s)e^{2\pi i\mathbf{s}\cdot\mathbf{r}_j}\sum_{\mathbf{k}}\mathbf{S}_{\mathbf{k}j}\sum_{\mathbf{H}}\delta(\mathbf{s}-\mathbf{k}-\mathbf{H}) \qquad (41)$$

The expression (41) indicates that the magnetic intensity is practically zero in the whole reciprocal space except at positions given by:

$$\mathbf{s} = \mathbf{h} = \mathbf{H}+\mathbf{k} \qquad (42)$$

Magnetic diffraction appears like a filter. Each satellite is decoupled of the rest of satellites, so if there are different propagation vectors **k** there is no interference between them, so there is always a phase factor between the Fourier coefficients $\mathbf{S_k}$ corresponding to different propagation vectors that is not accessible by diffraction methods unless a crystal structure distortion is coupled with the magnetic ordering. Notice that the concept of "fundamental reflections" (used in modulated crystal structures) does not apply here because **h**=**H** corresponds to nuclear reflections. Only when **k**=0 there is a magnetic contribution in top of the nuclear reflections.

For a particular magnetic reflection indexed as in (42) the magnetic structure factor of the unit cell is:

$$\mathbf{M}(\mathbf{h}) = \mathbf{M_h} = p\sum_{j} f_j(h)\mathbf{S}_{\mathbf{k}j}e^{2\pi i\mathbf{h}\cdot\mathbf{r}_j} = p\sum_{j} f_j(|\mathbf{H}+\mathbf{k}|)\mathbf{S}_{\mathbf{k}j}e^{2\pi i(\mathbf{H}+\mathbf{k})\cdot\mathbf{r}_j} \qquad (43)$$

The intensity of a magnetic Bragg reflection is proportional to the square of the magnetic interaction vector:

$$\mathbf{M}_{\perp\mathbf{h}} = \frac{1}{h^2}\mathbf{h}\times\mathbf{M_h}\times\mathbf{h} = \mathbf{e}\times\mathbf{M_h}\times\mathbf{e} = \mathbf{M_h}-(\mathbf{e}\cdot\mathbf{M_h})\mathbf{e} \qquad (44)$$

where **e** is the unit vector along the scattering vector **h**=**H**+**k**. In the case of a propagation vector **k**=**0** the intensity of a Bragg reflection for non polarised neutrons is given by:

$$I_{\mathbf{h}} = N_{\mathbf{h}}N_{\mathbf{h}}^* + \mathbf{M}_{\perp\mathbf{h}}\cdot\mathbf{M}_{\perp\mathbf{h}}^* \qquad (45)$$

where $N_{\mathbf{h}} = F(\mathbf{h})$ is the nuclear structure factor, otherwise only the second term (pure magnetic scattering) of the sum contributes to the intensity of reflection **h**.

7. The symmetry analysis based on representation theory for determining magnetic structures

Magnetic neutron diffraction experiments provide as immediate information the position of magnetic reflections in reciprocal space. This allows the determination of the propagation vector(s) characterising the magnetic moment configuration given by the Fourier series (34). The measurement of the intensities gives us a list of non-linear equations with the Fourier coefficients $\mathbf{S}_{\mathbf{k}j}$ as unknowns (equations 43-45). The problem of determining the magnetic structure of a crystalline solid, for which the crystal structure is known, reduces to the determination of the quantities $\{\mathbf{k}\}$ and $\{\mathbf{S}_{\mathbf{k}j}\}$ from which we can apply the expression (34) for determining the magnetic moment of whatever atom in the crystal. This result takes into account only the translational symmetry of the crystal. In general we have enough information from the whole symmetry of the crystal structure to further reduce the number of free independent parameters. Let us apply the general method of group representations (summarised in section 4) to the problem of finding and classifying the possible magnetic structures that can be realised in the frame of a particular crystal structure. Notice that in the forthcoming sections we will not use the time inversion symmetry (primed operators). The concept of propagation vector(s) in the Fourier series (34) replaces to some extend the "anti-translations" in the description of commensurate magnetic structures having $\mathbf{k}=1/2\ \mathbf{H}$ (see formula (36)).

7.1: *The working space for symmetry analysis of magnetic structures: magnetic representation*

Up to now we have considered the index *j* for labelling magnetic atoms in a primitive cell without taking into account the space group and the different Wyckoff positions. We do the analysis for the propagation vector $\mathbf{k}$. Let us now consider that the magnetic atom site *j* has equivalent atoms labelled as *js* ($j1$, $j2$, …jp_j) under the application of symmetry operators of the crystal space group $\mathbf{G}$ belonging also to the wave vector group $\mathbf{G}_{\mathbf{k}}$. The group $\mathbf{G}_{\mathbf{k}}$ is formed by the set of symmetry operators that leave invariant the propagation vector modulo a reciprocal lattice vector: $\mathbf{G}_{\mathbf{k}}=\{g\in\mathbf{G} \mid g\mathbf{k}=\mathbf{k}+\mathbf{H},\ \mathbf{H}\in L^*\}$, where L^* is the crystallographic reciprocal lattice. One can generate a reducible representation of $\mathbf{G}_{\mathbf{k}}$ by considering the complex working space spanned by all the components of $\mathbf{S}_{\mathbf{k}js}$. Each complex vector has three components (but two parameters per component: modulus and phase or real and imaginary part). As the atoms belonging to different sites do not mix under the symmetry operators, we can treat separately the different sites. For us the index *j* is then fixed and the index *s* varies from 1 to p_j. Being p_j the number of sublattices generated by the site *j*. The working complex space for site *j* has dimension $n_j=3\times p_j$ and it is spanned by unit vectors $\{\boldsymbol{\varepsilon}_{s\alpha}^{\mathbf{k}j}\}$ ($\alpha = 1, 2, 3$ −or *x, y, z* and $s = 1… p_j$,) represented as column vectors (with a single index *n*) with zeroes except for $n=\alpha+3(s-1)$ for which the component is one. The vectors $\{\boldsymbol{\varepsilon}_{s\alpha}^{\mathbf{k}j}\}$ are formed by direct sums (juxtaposition) of normal 3D unit vectors $\{\mathbf{u}_{s\alpha}^{\mathbf{k}j}\}$. For instance one of the vectors is given by:

$$\boldsymbol{\varepsilon}_{s\alpha}^{\mathbf{k}j} = \sum_{\oplus s} \mathbf{u}_{s\alpha}^{\mathbf{k}j} = \begin{pmatrix} \mathbf{u}_{1\alpha}^{\mathbf{k}j} \\ \mathbf{u}_{2\alpha}^{\mathbf{k}j} \\ \vdots \\ \mathbf{u}_{p_j\alpha}^{\mathbf{k}j} \end{pmatrix} = \begin{pmatrix} u_{1x}^{\mathbf{k}j} \\ u_{1y}^{\mathbf{k}j} \\ u_{1z}^{\mathbf{k}j} \\ u_{2x}^{\mathbf{k}j} \\ \vdots \\ \vdots \\ u_{p_jy}^{\mathbf{k}j} \\ u_{p_jz}^{\mathbf{k}j} \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 0 \\ \vdots \\ 1 \\ \vdots \\ 0 \end{pmatrix}$$

The vectors $\{ \boldsymbol{\varepsilon}_{s\alpha}^{\mathbf{k}j} \}$ may be considered as the columns of the unit matrix of dimension $n_j \times n_j = 9p_j^2$:

$$\{ \boldsymbol{\varepsilon}_{11}^{\mathbf{k}j} \quad \boldsymbol{\varepsilon}_{12}^{\mathbf{k}j} \quad \boldsymbol{\varepsilon}_{13}^{\mathbf{k}j} \quad \ldots \quad \ldots \quad \boldsymbol{\varepsilon}_{p_j2}^{\mathbf{k}j} \quad \boldsymbol{\varepsilon}_{p_j3}^{\mathbf{k}j} \} = \begin{pmatrix} 1 & 0 & 0 & \ldots & \ldots & 0 & 0 \\ 0 & 1 & 0 & \ldots & \ldots & 0 & 0 \\ 0 & 0 & 1 & \ldots & \ldots & 0 & 0 \\ \vdots & \vdots & \vdots & \ddots & \vdots & \vdots & \vdots \\ \vdots & \vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\ 0 & 0 & 0 & \ldots & \ldots & 1 & 0 \\ 0 & 0 & 0 & \ldots & \ldots & 0 & 1 \end{pmatrix}$$

The n_j $\{ \boldsymbol{\varepsilon}_{s\alpha}^{\mathbf{k}j} \}$ vectors refer to the zero-cell. One can extend the basis vectors to the whole crystal by using the Bloch propagation then forming column vectors of up to $n_j \times N = 3 \times p_j \times N$ dimensions, being N the total number of unit cells in the crystal. The component number n, for each kind of vector (3D, $3p_j$D and $3Np_j$D), is given by the expressions:

$$\boldsymbol{\varepsilon}_{s\alpha}^{\mathbf{k}j} = \sum_{\oplus s} \mathbf{u}_{s\alpha}^{\mathbf{k}j} \qquad 3D \rightarrow (\mathbf{u}_{s\alpha}^{\mathbf{k}j})_n = \delta_{n,\alpha}; \quad 3p_jD \rightarrow (\boldsymbol{\varepsilon}_{s\alpha}^{\mathbf{k}j})_n = \delta_{n,\alpha+3(s-1)}$$

$$\boldsymbol{\varphi}_{s\alpha}^{\mathbf{k}j} = \sum_{\oplus l} \boldsymbol{\varepsilon}_{s\alpha}^{\mathbf{k}j} \exp(-2\pi i \mathbf{k}\mathbf{R}_l) \qquad 3Np_jD \rightarrow (\boldsymbol{\varphi}_{s\alpha}^{\mathbf{k}j})_n = e^{-2\pi i \mathbf{k}\mathbf{R}_l} \delta_{n,\alpha+3(s-1)+3lp_j} \tag{46}$$

If one applies the symmetry operators of $\mathbf{G_k}$ to the vectors $\{ \boldsymbol{\varepsilon}_{s\alpha}^{\mathbf{k}j} \}$ taking into account that they are axial vectors we obtain another vector (after correcting for the Bloch phase factor if the operator moves the atom outside the reference zero-cell) of the same basis. The matrices $\Gamma_{s\alpha,q\beta}^{\mathbf{k}j}(g)$ of dimension $n_j \times n_j$ corresponding to the different operators constitute what is called the "Magnetic Representation" for the site j and propagation vector $\mathbf{k}$. Let us get an explicit form for these matrices.

The action of the operator $O(g)$ on the vector $\{ \boldsymbol{\varepsilon}_{s\alpha}^{\mathbf{k}j} \}$ can be obtained from the equations (3) and (4) but considering only conventional crystallographic operators. Applying a symmetry operator to the vector position and the unit spin associated to the atom js along the α-axis, changes the index js to jq and reorient the spin according to the nature of the operator $g=\{h|\mathbf{t}_h\}$ for axial vectors.

$$g\mathbf{r}_s^j = h\mathbf{r}_s^j + \mathbf{t}_h = \mathbf{r}_q^j + \mathbf{a}_{gs}^j; \quad gs \rightarrow (q, \mathbf{a}_{gs}^j)$$

$$O(g)\mathbf{u}_{s\alpha}^{\mathbf{k}j} = \det(h) \, h\mathbf{u}_{gs,\alpha}^{\mathbf{k}j} = \det(h) \sum_m h_{\beta m} (\mathbf{u}_{q\alpha}^{\mathbf{k}j})_m = \det(h) \sum_m h_{\beta m} \delta^q_{m,\alpha} = \det(h) h_{\beta\alpha}[q] \tag{47}$$

The operator g acting on atom positions permutes the numbering of the atoms belonging to a same site (orbit) and provides a returning vector $\mathbf{a}_{gs}^j$, which must appear in a phase factor when working with Bloch functions when the transformed atom is outside the zero-cell.

$$O(g)\boldsymbol{\varepsilon}_{s\alpha}^{\mathbf{k}j} = \sum_{q\beta} \Gamma_{q\beta,s\alpha}^{\mathbf{k}j}(g)\boldsymbol{\varepsilon}_{q\beta}^{\mathbf{k}j} = \sum_{q\beta} e^{2\pi i \mathbf{k}\mathbf{a}_{gs}^j} \det(h)h_{\beta\alpha}\delta_{s,gq}^j \boldsymbol{\varepsilon}_{q\beta}^{\mathbf{k}j} \tag{48}$$

The explicit components for the magnetic representation are:

$$\Gamma_{Mag} \rightarrow \Gamma_{q\beta,s\alpha}^{\mathbf{k}j}(g) = e^{2\pi i \mathbf{k}\mathbf{a}_{gs}^j} \det(h)h_{\beta\alpha}\delta_{q,gs}^j \tag{49}$$

The δ symbol has value equal to 1 when the operator g transforms the atom s into the atom q, and zero otherwise.

7.2: *The magnetic representation as direct product of permutation and vector representation*

An inspection to the explicit expression for the magnetic representation, given by equation (49), for the propagation vector $\mathbf{k}$, the Wyckoff position j, with sublattices indexed by (s, q), shows that it may be considered as the direct product of the so called "permutation representation" by the axial (or in general "vector") representation. The explicit matrices of the permutation representation, of dimension $p_j \times p_j$, are:

$$\Gamma_{Perm} \rightarrow P_{qs}^{\mathbf{k}j}(g) = e^{2\pi i \mathbf{k}\mathbf{a}_{gs}^j}\delta_{q,gs}^j \tag{50}$$

The matrices of the axial representation, of dimension 3, are constituted by the rotational part of the $\mathbf{G_k}$ operators multiplied by -1 when the operator $g=\{h|\mathbf{t}_h\}$ corresponds to an improper rotation.

$$\Gamma_{Axial} \rightarrow V_{\beta\alpha}(g) = \det(h)h_{\beta\alpha} \tag{51}$$

$$\Gamma_{Mag} = \Gamma_{Perm} \otimes \Gamma_{Axial}$$

The magnetic representation Γ_{Mag}, irrespective of the indices, can be decomposed in irreducible representations of the propagation vector group. Using the expressions (9-12) and taking into account that, though the space group is infinite, we work only with the coset representatives with respect to the translation subgroup, we can calculate *a priori* the number of possible basis functions of the *irreps* of $\mathbf{G_k}$ that describe the possible magnetic structures. This number (N_{BV}) is equal to the number of times the representation Γ^ν is contained in Γ_{Mag} times the dimension of Γ^ν: $N_{BV}=n_\nu \times l_\nu$.

7.3: *Projection operators and basis vectors of irreducible representations of $\mathbf{G_k}$*

The basis functions of the *irreps* of $\mathbf{G_k}$ can be calculated using the projection operator formula (17) particularised for the explicit expression of $O(g)$ acting on the vectors $\{\boldsymbol{\varepsilon}^{\mathbf{k}j}_{\alpha s}\}$ (equation 48). The explicit formula giving the n_j-dimensional basis vectors of the representation Γ^ν for site j is the following:

$$\psi_\lambda^{\mathbf{k}\nu}(j) = \frac{1}{n(\mathbf{G_{0k}})} \sum_{g \in \mathbf{G_{0k}}} \Gamma_{\lambda[\mu]}^{*\nu}(g)\, O(g)\, \boldsymbol{\varepsilon}_{s\alpha}^{\mathbf{k}j} \qquad\qquad (\lambda = 1,...l_\nu)$$

$$\psi_\lambda^{\mathbf{k}\nu}(j) = \frac{1}{n(\mathbf{G_{0k}})} \sum_{g \in \mathbf{G_{0k}}} \Gamma_{\lambda[\mu]}^{*\nu}(g) \sum_{q\beta} \exp(2\pi i\, \mathbf{k}\, \mathbf{a}_{gs}^j)\det(h)h_{\beta\alpha}\delta_{s,gq}^j \boldsymbol{\varepsilon}_{q\beta}^{\mathbf{k}j}$$

(52)

It is convenient to use, instead of the basis vectors for the whole set of magnetic atoms in the primitive cell, the so called *atomic components* of the basis vectors, which are normal 3D constant vectors attached to individual atoms:

$$\psi_\lambda^{\mathbf{k}\nu}(j) = \sum_{\oplus,\,s=1,...p_j} \mathbf{S}_\lambda^{\mathbf{k}\nu}(js)$$

(53)

The explicit expression for the atomic components of the basis functions is:

$$\mathbf{S}_\lambda^{\mathbf{k}\nu}(js) \propto \sum_{g \in \mathbf{G_{0k}}} \Gamma_{\lambda[\mu]}^{*\nu}(g)\, e^{2\pi i\, \mathbf{k}\, \mathbf{a}_{gs}^j}\det(h)\delta_{s,g[q]}^j \begin{pmatrix} h_{1\alpha} \\ h_{2\alpha} \\ h_{3\alpha} \end{pmatrix}$$

(54)

The possible magnetic structures, described by Fourier coefficients, can be obtained in terms of hopefully less number of independent parameters if these Fourier components are linear combinations of the basis vectors (54).

We have considered up to now a single **k**-vector and the representations of the wave vector group $\mathbf{G_k}$. It is convenient to derive the basis vectors corresponding to the full star in terms of the basis vectors of a single arm. If we assume that the transition involves all the arms of the star (representations of the full space group) we can eventually obtain more constraints and reduce the number of free parameters. Taking into account the fact that for the arm $\mathbf{k}_L = g_L\mathbf{k}$ (see equations (27)) the wave vector group is conjugate to $\mathbf{G_k}$ ($\mathbf{G_{kL}} = g_L\, \mathbf{G_k}\, g_L^{-1}$) we obtain the following relation between the atomic components of the basis vectors:

$$\mathbf{S}_\lambda^{\mathbf{k}_L\nu}(js) = e^{2\pi i\, \mathbf{k}_L\, \mathbf{a}_{g_L q}^j}\det(h_L)h_L \mathbf{S}_\lambda^{\mathbf{k}\nu}(jq)$$

(55)

The atoms s and q of the site j and the returning vector given in the exponential term are related by the equation:

$$g_L \mathbf{r}_q^j = h_L \mathbf{r}_q^j + \mathbf{t}_{h_L} = \mathbf{r}_s^j + \mathbf{a}_{g_L q}^j$$

(56)

Notice that g_L is an operator of the full space group that connects atoms that may be split into different orbits in the wave vector group $\mathbf{G_k}$. We can impose further constraints by applying the relation (55) not just to basis vectors but to Fourier vector coefficients belonging to different orbits reducing the number of free parameters to describe a magnetic structure.

The so-called multi-**k** structures correspond to magnetic structures in which the full set of star-arms participates in the spin configuration. It is very difficult to determine how many arms, *transition channel* [15], are contributing to an experimental magnetic structure (see chapter 4, starting at section 18, of reference [15]) unless a physical test can be performed to distinguish a real multi-**k** structure from the presence of magnetic domains.

A general magnetic structure can be described by a Fourier series as (34) where the index j was for a generic magnetic atom irrespective of symmetry. The Fourier series can be rewritten using the notation used in the present section, simply by changing the index j for the double index js. The fundamental hypothesis of the symmetry analysis, when working with magnetic structures, is that the Fourier coefficients $\mathbf{S}_{\mathbf{k}js}$ that describe magnetic structures with propagation vector $\mathbf{k}$ must be a linear combination of the basis functions (atomic components) of the $\mathbf{G}_{\mathbf{k}}$ irreducible representations.

$$\mathbf{S}_{\mathbf{k}js} = \sum_{n\lambda} C_{n\lambda}^{\mathbf{k}\nu} \mathbf{S}_{n\lambda}^{\mathbf{k}\,\nu}(js) \tag{57}$$

where ν labels the active irreducible representation, Γ^ν, of the propagation vector group $\mathbf{G}_{\mathbf{k}}$, λ labels the component corresponding to the dimension of the representation Γ^ν, n is an additional index (with respect to expression (54)) running between one and the number of times ($n=1...n_\nu$) the representation Γ^ν is contained in the global magnetic representation Γ_{Mag}. The quantities $\mathbf{S}_{n\lambda}^{\mathbf{k}\,\nu}(js)$ are constant vectors, in general complex, obtained by the application of the projection operator formula to axial unit vectors along the directions of the unit cell axes attached to the positions js. The explicit formula for obtaining them is the equation (54). These vectors are similar to the polarisation vectors of normal modes in lattice dynamics except that they are axial vectors. The basis vectors may be normalised in different ways. An additional sum over ν is sometimes necessary when more than one irreducible representation is involved in the magnetic phase transition (see reference [15] for examples and details). In the case the representation analysis is fully used, the mixing coefficients $C_{n\lambda}^{\mathbf{k}\nu}$ are the free parameters of the magnetic structure (they correspond to the *order parameters* in the Landau theory of phase transitions) and usually the total number is lower than the number of Fourier components of each magnetic atom in the unit cell. If we add a sum over representations, the expression (57) gives the most general case; in practice one has to assume additional constraints because the number of mixing coefficients may be too high. In particular for multidimensional representations one can select a particular direction in the representation space (for instance not considering all the λ-indices) in order to diminish the number of parameters. In the case of commensurate magnetic structures this selection corresponds to the assumption of different Shubnikov groups. The same may be stated for incommensurate structures when superspace groups are considered.

The number of free coefficients to describe a magnetic structure corresponding to a single representation of $\mathbf{G}_{\mathbf{k}}$ is related to the number of independent basis vectors $n_f \propto n_\nu \times \dim(\Gamma^\nu)$. In the general case the basis functions can be complex vectors as well as the coefficients $C_{n\lambda}^{\mathbf{k}\nu}$, the condition $\mathbf{S}_{\mathbf{k}js} = \mathbf{S}_{-\mathbf{k}js}^*$ assures the reality of the magnetic moments. The effective number of free parameters depends on some additional assumptions related to the consideration of the star of the wave vector. Let us consider only a single wave vector and the representations of $\mathbf{G}_{\mathbf{k}}$. In such a case, the analysis is successful when the number of free parameters is lower than $6p_j$ in the case of $\mathbf{k}$ not equivalent to $-\mathbf{k}$ or $3p_j$ in case of real Fourier coefficients. In summary, the group theory, considering only $\mathbf{G}_{\mathbf{k}}$, is useful when:

$$n_f = 2\,n_\nu \times \dim(\Gamma^\nu) < 6\,p_j \quad \text{(for } \mathbf{k} \text{ non equivalent to } -\mathbf{k}) \tag{58}$$

$$n_f = \quad n_\nu \times \dim(\Gamma^\nu) < 3\,p_j \quad \text{(for } \mathbf{k} \text{ equivalent to } -\mathbf{k}) \tag{58'}$$

The factor 2 comes from the fact that, in general, the coefficients may be complex (modulus and phase). When the constraints introduced by symmetry analysis are not enough to simplify the problem and tackle successfully the experimental data, one has to consider other kind of restrictions imposed by the previous knowledge of the physics of the system. The most common constraint, based on physical grounds, is that the magnetic moment of the different atoms belonging to a same crystallographic site should have the same modulus, at least for commensurate magnetic structures.

Let us consider briefly the invariance properties of a magnetic structure described by the propagation vector $\mathbf{k}$. Using the full basis vectors of a single Wyckoff site of $\mathbf{G_k}$ (direct sum over $s=1...p$, see eq. (53)) we can remove the index s and write:

$$\mathbf{S_{ks}} = \sum_{n\lambda} C_{n\lambda}^{\mathbf{k}\nu} \mathbf{S}_{n\lambda}^{\mathbf{k}\nu}(s) \quad \Rightarrow \quad \mathbf{S_{[k]}} = \sum_{n\lambda} C_{n\lambda}^{\mathbf{k}\nu} \boldsymbol{\psi}_{n\lambda}^{\mathbf{k}\nu} \tag{59}$$

If an operator g of $\mathbf{G_k}$ is a symmetry operator of the magnetic structure it must verify[4]:

$$O(g)\mathbf{S_{[k]}} = \mathbf{S_{[k]}} \tag{60}$$

$$O(g)\mathbf{S_{[k]}} = \sum_{n\lambda} C_{n\lambda}^{\mathbf{k}\nu} O(g)\boldsymbol{\psi}_{n\lambda}^{\mathbf{k}\nu} = \sum_{n\lambda} C_{n\lambda}^{\mathbf{k}\nu} \sum_{\lambda'} \Gamma_{\lambda'\lambda}^{\mathbf{k}\nu}(g)\boldsymbol{\psi}_{n\lambda'}^{\mathbf{k}\nu} = \sum_{n\lambda'} \sum_{\lambda} \Gamma_{\lambda'\lambda}^{\mathbf{k}\nu}(g)C_{n\lambda}^{\mathbf{k}\nu}\boldsymbol{\psi}_{n\lambda'}^{\mathbf{k}\nu} = \sum_{n\lambda'} C_{n\lambda'}^{\mathbf{k}\nu}\boldsymbol{\psi}_{n\lambda'}^{\mathbf{k}\nu} \tag{60'}$$

So that,

$$C_{n\lambda'}^{\mathbf{k}\nu} = \sum_{\lambda} \Gamma_{\lambda'\lambda}^{\mathbf{k}\nu}(g)C_{n\lambda}^{\mathbf{k}\nu} \tag{61}$$

where $\Gamma_{\lambda'\lambda}^{\mathbf{k}\nu}(g)$ are the matrix elements of the *irrep* $\Gamma^{\mathbf{k}\nu}$ of $\mathbf{G_k}$ corresponding to the operator g. If g is a symmetry operator of the magnetic structure, then the mixing coefficients $C_{n\lambda}^{\mathbf{k}\nu}$ must satisfy the equation (61).

We can extend the same considerations to a magnetic structure described by the full set of arms of $\{\mathbf{k}\}$. If we consider a single Wyckoff site of the full space group, the Fourier coefficients and the basis vectors of the full *irrep* $\Gamma^{\{\mathbf{k}\}\nu}$ can be written and related as:

$$\mathbf{S_{[\{k\}]}} = \sum_{L} \sum_{n\lambda} C_{n\lambda}^{\mathbf{k}_L\nu} \boldsymbol{\psi}_{n\lambda}^{\mathbf{k}_L\nu}; \quad O(g)\mathbf{S_{[\{k\}]}} = \mathbf{S_{[\{k\}]}} \Rightarrow C_{n\lambda}^{\mathbf{k}_L\nu} = \sum_{L'} \sum_{\lambda'} \Gamma_{L\lambda,L'\lambda'}^{\{\mathbf{k}\}\nu}(g)C_{n\lambda'}^{\mathbf{k}_{L'}\nu} \tag{62}$$

In this case the matrix elements $\Gamma_{L\lambda,L'\lambda'}^{\{\mathbf{k}\}\nu}(g)$ correspond to those of a full irreducible representation of the space group $\mathbf{G}$. They are related to the *irreps* matrices of $\mathbf{G_k}$ by the induction formula (see eq. 30). The relations (62) put the appropriate constraints between mixing coefficients in order to assume that the operator $g \in \mathbf{G}$ (or $g \in \mathbf{G_P}$) is a symmetry operator of the magnetic structure.

For incommensurate magnetic structures one can extend the invariance equation (62) by including a global phase factor associated to the symmetry operator for each pair of $(\mathbf{k}, -\mathbf{k})$ arms of the star:

$$O(g)\mathbf{S_{[k, -k]}} = \mathbf{S_{[k, -k]}}\, e^{-2\pi i\varphi_g} \tag{62'}$$

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[4] For the invariance symmetry we have to consider also operators combined with time inversion. We can use the representations of the paramagnetic group ($\mathbf{G_P}=\mathbf{G}+\mathbf{G}1'$), which have a one to one correspondence to those of the space group $\mathbf{G}$ without need of using co-representations (useful in quantum mechanics): the matrices corresponding to primed elements are just the product of those corresponding to unprimed elements by the negative identity matrix. Symbolically $\Gamma(g') = -\Gamma(g)$, with g, g'$\in \mathbf{G_P}$.

This extends the symmetry invariance to incommensurate structures. The full treatment should be done using the superspace formalism not treated in this document.

For the case of incommensurate magnetic structures one has to consider at least two wave vectors in the arm: $\mathbf{k}$ and $-\mathbf{k}$. In such cases it may be possible that atoms belonging to a same orbit in the full space group are split into two orbits in $\mathbf{G_k}$. The atomic components of the basis vectors of the atoms belonging to the two orbits are related by the relations (55). When using the superposition (57) for $\mathbf{k}$ and $-\mathbf{k}$ and the condition $\mathbf{S}_{\mathbf{k}js} = \mathbf{S}^*_{-\mathbf{k}js}$, one can obtain the relations between the coefficients $C^{\mathbf{k}\nu}_{n\lambda}$ and $C^{-\mathbf{k}\nu}_{n\lambda}$. If we assume additional symmetry constraints by using the relations (62), or the equation (55) applied to $\mathbf{S}_{\mathbf{k}js}$, restricted to the pair $(\mathbf{k}, -\mathbf{k})$, we can fix the relation between the Fourier coefficients of the two orbits.

Instead of using directly the mixing coefficients for describing a magnetic structure, one can use a more traditional crystallographic approach in some cases. The Fourier component $\mathbf{k}$ of the magnetic moment of atom $j1$, which transforms to the atom js when the symmetry operator $g_s = \{h|\mathbf{t}\}_s$ of $\mathbf{G_k}$ is applied ($\mathbf{r}^j_s = g_s\mathbf{r}^j_1 = h_s\mathbf{r}^j_1 + \mathbf{t}_s$), is transformed as:

$$\mathbf{S}_{\mathbf{k}js} = M_{js}\mathbf{S}_{\mathbf{k}j1}exp\{-2\pi i\phi_{\mathbf{k}js}\} \tag{63}$$

The matrices M_{js} and phases $\phi_{\mathbf{k}js}$ can be deduced from the relations between the Fourier coefficients and atomic basis functions (57). The matrices M_{js} correspond, in the case of commensurate magnetic structures, to the rotational parts of the magnetic Shubnikov group acting on magnetic moments.

7.5: *Magnetic Space Groups and Irreducible Representations of dimension 1*

There exists a well defined relation between the theory of magnetic space groups (Shubnikov groups) and the representation theory. This is known in the literature as the Niggli-Indenbom theorem and it is related to the fundamental lemma discussed in section 2 about the subgroups of index 2 of crystallographic groups. The theorem states that: *the magnetic space groups correspond to real 1D irreducible representations of conventional crystallographic space groups*. This is easily seen by considering the following: suppose we have a real representation Γ of the space group $\mathbf{G}$, the matrices of this representation can be of the form $\Gamma(g)=1$ or $\Gamma(f)=-1$; if we associate the time reversal operator $1'$ to the elements $f \in \mathbf{G}$, so they become primed elements f', the multiplication table of g-type and f'-type elements coincides with the multiplication table of the $\{\Gamma(g) ..., \Gamma(f)...\}$ matrices. There is then an isomorphism between magnetic groups and real 1-dimensional irreducible representations of space groups.

8. The magnetic structure factor when symmetry is considered.

If isotropic thermal motion is considered and if symmetry relations are established for coupling the different Fourier components, we obtain the general expression of the magnetic structure factor:

$$\mathbf{M(h)} = p\sum_{j=1}^{n_a}O_jf_j(\mathbf{h})e^{-B_j|h/2|^2}\sum_{s=1,...p_j}M_{js}\mathbf{S}_{\mathbf{k}j1}exp\{2\pi i[(\mathbf{H}+\mathbf{k})\{h|\mathbf{t}\}_s\mathbf{r}^j_1 - \psi_{\mathbf{k}js}]\} \tag{64}$$

The sum over j concerns the atoms of the magnetic asymmetric unit for the wave vector $\mathbf{k}$. So that j labels different sites. The sum over s concerns the symmetry operators of the wave vector group $\mathbf{G_k}$. The phase factor $\psi_{\mathbf{k}js}$ has two components:

$$\psi_{\mathbf{k}js} = \Phi_{\mathbf{k}j} + \phi_{\mathbf{k}js} \tag{65}$$

$\Phi_{\mathbf{k}j}$ is a phase factor that is not determined by symmetry. It is a free parameter and it is significant only for an independent set of magnetic atoms (one orbit) which respect to another one. The component $\phi_{\mathbf{k}js}$ is a phase factor determined by symmetry as shown in (63). The sign of $\phi_{\mathbf{k}js}$ changes for $-\mathbf{k}$. In the general case $\mathbf{S}_{\mathbf{k}j1}$ is a complex vector with six components. These six components per magnetic orbit constitute the parameters that have to be refined from the diffraction data. Symmetry reduces hopefully the number of free parameters per orbit to be refined. Notice that we have adopted a different phase sign convention than that used in reference [19].

In the case of a commensurate magnetic structure one can calculate the magnetic structure factor in the magnetic unit cell. In such a case $\mathbf{S}_{\mathbf{k}j1}$ are real vectors corresponding to the magnetic moment of the atom $j1$, the matrices M_{js} are all real and the phases verify $\phi_{\mathbf{k}js} = 0$. The crystallographic magnetic group theory can be fully applied in such a case [1-5].

In the case the general decomposition in terms of atomic basis functions (57) is used, the magnetic structure factor can be written as:

$$\mathbf{M}(\mathbf{h}) = p\sum_{j=1}^{n_a} O_j f_j(\mathbf{h}) e^{-B_j |h/2|^2} \sum_{n\lambda} C_{n\lambda}^{\mathbf{k}\nu} \sum_s \mathbf{S}_{n\lambda}^{\mathbf{k}\,\nu}(js) exp\{2\pi i (\mathbf{h}_s \mathbf{x}_j + \mathbf{h} \cdot \mathbf{t}_s)\} \tag{66}$$

where we have written $\mathbf{h}_s = h_s^T \mathbf{h}$ (superscript T stands here for transpose)

9. Limitations of neutron scattering for determining magnetic structures

If the magnetic structure has several propagation vectors $\mathbf{k}$, and there is no strong coupling with the crystal lattice, it is not possible to determine unambiguously the spin configuration because the phase between the different Fourier components cannot be determined by diffraction methods. Here we take j as the index of a generic magnetic atom (irrespective of symmetry sites). One can see easily what the origin of this problem is; let us rewrite the expression (34) as:

$$\mathbf{m}_{lj} = \sum_{\mathbf{k}} \mathbf{S}_{\mathbf{k}j} exp\{-2\pi i(\mathbf{k}\mathbf{R}_l + \Phi_{\mathbf{k}})\} = \sum_{\mathbf{k}} \mathbf{S}_{\mathbf{k}j}^m exp\{-2\pi i\mathbf{k}\mathbf{R}_l\} \tag{67}$$

In which we have added an arbitrary phase factor $\Phi_{\mathbf{k}}$ depending only on $\mathbf{k}$. We can understand easily that the modified Fourier coefficients, $\mathbf{S}_{\mathbf{k}j}^m$, give rise, in general, to another kind of magnetic structure specially if the structure is commensurate. However, the diffraction pattern obtained with a Fourier series like (67) is identical to that obtained with (34). This is readily demonstrated because the modified magnetic structure factors (43) and the intensity corresponding to the reflection $\mathbf{h}=\mathbf{H}+\mathbf{k}$ are given by:

$$\mathbf{M}^m(\mathbf{h}) = p\sum_j f_j(h)\mathbf{S}_{\mathbf{k}j}^m e^{2\pi i(\mathbf{H}+\mathbf{k})\cdot\mathbf{r}_j} = pe^{-2\pi i\Phi_{\mathbf{k}}}\sum_j f_j(h)\mathbf{S}_{\mathbf{k}j}e^{2\pi i(\mathbf{H}+\mathbf{k})\cdot\mathbf{r}_j} = e^{-2\pi i\Phi_{\mathbf{k}}}\mathbf{M}(\mathbf{h})$$

$$\mathbf{I}_{mag}(\mathbf{h}) = \mathbf{M}_{\perp}^m(\mathbf{h}) \cdot \mathbf{M}_{\perp}^{*m}(\mathbf{h}) = e^{-2\pi i\Phi_{\mathbf{k}}}e^{+2\pi i\Phi_{\mathbf{k}}}\mathbf{M}_{\perp}(\mathbf{h}) \cdot \mathbf{M}_{\perp}^*(\mathbf{h}) = \mathbf{M}_{\perp}(\mathbf{h}) \cdot \mathbf{M}_{\perp}^*(\mathbf{h}) \tag{68}$$

The arbitrary phase factor disappears when calculating the intensity. So, if several propagation vectors exist in a diffraction pattern, infinitely many structures are able to explain the observed pattern and diffraction alone is unable to provide a unique solution. Symmetry constrains and, more importantly, restrictions on the amplitude of the magnetic moments can reduce the number of solutions. A study of the different kinds of propagation vectors that can be combined in order to provide constant moment structures, among the study of the physical properties, was performed long time ago by Nagamiya [20]. Fortunately, nature often selects simple solutions and many magnetic structures have a single propagation vector, or display some symmetry constraints that reduce the complexity of the periodic magnetic structures given by (34).

Special consideration requires the sinusoidal structures in which the single pair ($\mathbf{k}$,-$\mathbf{k}$) of propagation vectors at the interior of the Brillouin Zone has commensurate components. Here the change of the global phase $\Phi_{\mathbf{k}}$ modifies the physical properties of the magnetic arrangement. If $\mathbf{k}$ is incommensurate the change of phase means only a change of global origin in the crystal and all the amplitudes of the magnetic moments between extreme values are realized somewhere in the crystal. If $\mathbf{k}$ is commensurate some particular values of the phases give a picture of the magnetic ordering that is really different of a conventional sinusoid. Let us consider the simplest case of a single atom per primitive unit cell and a propagation vector $\mathbf{k}$=1/4$\mathbf{H}$. To be specific, consider the case described in Figure 4(a). The magnetic moment, expression (67), in the lattice position $\mathbf{R}_l =(l_1, l_2, l_3)$ is given by:

$$\mathbf{m}_l = \sum_{\mathbf{k},-\mathbf{k}} \mathbf{S}_{\mathbf{k}} exp\{-2\pi i(\mathbf{k}\mathbf{R}_l + \Phi_{\mathbf{k}})\} = (0,0,w)\cos 2\pi(1/4\mathbf{H}\mathbf{R}_l + \Phi_{\mathbf{k}}) = (0,0,w)\cos 2\pi(\frac{l_2}{4} + \Phi_{\mathbf{k}})$$

with l_2 integer.

Two interesting cases occur:

   i)   If $\Phi_{\mathbf{k}} = 0$ the sequence of magnetic moment components along $\mathbf{c}$ for the lattice points l_2= (0, 1, 2, 3, 4, 5 …) are: (w, 0, -w, 0, w, 0 …).
   ii)   If $\Phi_{\mathbf{k}} = 1/8$ the sequence is: (w', w', -w', -w', w', w', -w', -w'…), with $w' = w/\sqrt{2}$.

Both sequences give exactly the same diffraction pattern up to the constant factor $1/\sqrt{2}$, so they are indistinguishable. In the first case there are paramagnetic atoms and in the second we have a constant moment magnetic structure. If the coupling of the magnetic moments with the crystal lattice is strong, one can distinguish the different solutions by their effect on the atomic displacements. It is clear that the pattern of atomic displacements in the case of $\Phi_{\mathbf{k}} = 0$ should be totally different from that of $\Phi_{\mathbf{k}} = 1/8$. In the major part of the cases the coupling is not strong enough to see nuclear superstructure reflections and we need other experimental techniques (Mössbauer spectroscopy, μ-SR, NMR …) that can help in getting the good choice among a set of indistinguishable solutions.

The problem of the degeneracy of solutions (different magnetic structures giving rise to the same diffraction pattern) is exacerbated in the case of powder diffraction. To a particular observed peak there is the contribution of different Bragg reflections so that the magnetic structure of compounds with symmetry higher than orthorhombic cannot be determined unambiguously. The paper by G. Shirane [21] examines the case of "uniaxial" (collinear)

magnetic structures and concludes that for cubic symmetry the direction of the magnetic moments cannot be determined by powder diffraction. In the case of tetragonal, rhombohedral and hexagonal systems, only the assumed "common" angle of the magnetic moments with the **c**-axis can be determined. The powder diffraction pattern is not sensitive to the orientation angle in the **a**-**b** plane.

10. Examples of commensurate magnetic structures with propagation vector at the centre or at the surface of the Brillouin Zone (BZ).

Let us consider the two types of magnetic structures described in (a) and (b) of section 5. These are respectively (a) **k**=**0** and (b) **k**=1/2**H**, where **H** is a reciprocal lattice vector (Liftchitz points of the BZ). Of course, the first type (a) is a particular case of the second (b) when **H**=0. We have separated the two cases only for signalling the largely spread confusion that when **k**=0, the magnetic structure is necessarily ferromagnetic. We shall consider a single orbit in the following paragraphs, so the index s labels the different sublattices of a single site.

For all magnetic structures with **k**=1/2**H** (now including **H**=0), the Fourier coefficients $\mathbf{S}_{\mathbf{k}j}$ should be real and there are cases where it is not possible to obtain from representation analysis real basis functions $\mathbf{S}_{n\lambda}^{\mathbf{k}\nu}s$ or a coherent set of coefficients $C_{n\lambda}^{\mathbf{k}\nu}$ allowing the description of a real magnetic structure. There are combinations of space groups and propagation vectors of the **k**=1/2**H** type giving rise to complex representations providing complex basis functions. Classically the Frobenius-Schur criterion for determining the type of representation (real or complex) for point groups can be extended for space groups (Herring's criterion). In summary we have the following cases:

$$\eta = \frac{l_k}{n(\mathbf{G}_{0\mathbf{k}})} \sum_{\substack{g=\{h|\mathbf{t}_h\}: \\ h\mathbf{k}=-\mathbf{k}+\mathbf{H}}} \chi^{\mathbf{k}\nu}(g^2) = \begin{cases} 1: & \Gamma & \textit{is real} \\ 0: & \Gamma \,\&\, \Gamma^* & \textit{are complex and non equivalent} \\ -1: & \Gamma \,\&\, \Gamma^* & \textit{are complex and equivalent} \end{cases} \quad (69)$$

where the sum is extended only for those operators of the space group transforming **k** into –**k** and l_k is the number of arms of the star of **k**. For the second and third cases the mixing of two or more irreducible representations is needed for obtaining real basis functions. In some cases the combination of several irreducible representations provides real basis functions but non-constant magnetic moments. One has to decide if for the particular physical situation this is an acceptable issue. In case the physical situation indicates a clear preference for constant magnetic moment it is necessary to reduce the symmetry of the initial space group and apply extra constraints. Let us consider four examples of the above general problem.

10.1: *Symmetry analysis of the magnetic structure of* LaMnO$_3$.

This compound contains a single Mn site and it is an example of the simplest case of a propagation vector **k**=0 and only real 1D *irreps* [22]. The space group is **G**=*Pbnm*, the cell parameters are a=5.695 Å, b=5.695 Å and c= 7.540 Å (see chapter by Grenier &Ballou). The Mn atoms are in the Wyckoff positions *4b*: 1(1/2,0,0), 2(1/2,0,1/2), 3(0,1/2,1/2) and 4(0,1/2,0). As the propagation vector is **k**=(0, 0, 0), the magnetic unit cell is identical to the nuclear cell. The whole symmetry analysis may be performed by hand [23] or by using one of

the available computing programs doing the work automatically [24-26]. We use hereafter the program BASIREPS [24].

The propagation vector is invariant, so $\mathbf{G_k}=\mathbf{G}=Pbnm$. The list of the irreducible representations (all of them of dimension 1) is given in Table 1, in which p=1/2.

Table 1: Irreducible representations of $\mathbf{G_k}=\mathbf{G}=Pbnm$.

```
----------------------------------------------------------------------------------
Irreps  {1}   {2_z|00p}  {2_x|pp0}   {2_y|ppp}   {-1}  m={m_z|00p}  b={m_x|pp0} n={m_y|ppp}  SG
----------------------------------------------------------------------------------
```

Irreps	{1}	{2_z|00p}	{2_x|pp0}	{2_y|ppp}	{-1}	m={m_z|00p}	b={m_x|pp0}	n={m_y|ppp}	SG
Γ_1:	1	1	1	1	1	1	1	1	*Pbnm*
Γ_2:	1	1	1	1	-1	-1	-1	-1	*Pb'n'm'*
Γ_3:	1	1	-1	-1	1	1	-1	-1	*Pb'n'm*
Γ_4:	1	1	-1	-1	-1	-1	1	1	*Pbnm'*
Γ_5:	1	-1	1	-1	1	-1	1	-1	*Pbn'm'*
Γ_6:	1	-1	1	-1	-1	1	-1	1	*Pb'nm*
Γ_7:	1	-1	-1	1	1	-1	-1	1	*Pb'nm'*
Γ_8:	1	-1	-1	1	-1	1	1	-1	*Pbn'm*

We have provided the symbol of the Shubnikov group (SG) corresponding to each irreducible representation (see sections 3 and 7.5) in the last column of the table. All *irreps* are real, so we are in the first case or rule (69).

We do not give the matrices of the magnetic representation Γ_m, of dimension $3\times p_j=3\times4 = 12$, because it can be easily deduced by hand or from the information given in the output file of BASIREPS. Γ_m has the characters: $\chi(\Gamma_m-4b) = $ (12, 0, 0, 0, 12, 0, 0, 0) and decomposes (see equations (9) and (12)) in terms of the *irreps* of the previous table as follows:

$$\Gamma_m(4b) = 3\Gamma_1 \oplus 3\Gamma_3 \oplus 3\Gamma_5 \oplus 3\Gamma_7$$

This means that if a single *irrep* defines the magnetic structure we have only three free parameters: $n_f= n_v\times dim(\Gamma_v)=3\times1=3$, which is well below the 12 components of magnetic moments in the primitive cell. The calculation of the basis vectors applying the formula (54) is done by BASIREPS, here we reproduce the output of the calculation for the *irrep* Γ_3 corresponding to the experimental magnetic structure:

```
+++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++
  Basis functions of Representation IRrep( 3) of dimension  1 contained 3 times in GAMMA
+++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++
                SYMM  x,y,z   -x+1,-y,z+1/2    -x+1/2,y+1/2,-z+1/2   x-1/2,-y+1/2,-z
                Atoms:      Mn_1            Mn_2                Mn_3             Mn_4
BsV( 1, 1: 4):Re (    1    0    0) (   -1    0    0) (    1    0    0) (   -1    0    0)
BsV( 2, 1: 4):Re (    0    1    0) (    0   -1    0) (    0   -1    0) (    0    1    0)
BsV( 3, 1: 4):Re (    0    0    1) (    0    0    1) (    0    0    1) (    0    0    1)

 The Fourier coefficients are LINEAR COMBINATIONS of Basis Functions: coefficients u,v,w,p,q…
 The general expressions of the Fourier coefficients Sk(j) of the atoms non-related
 by lattice translations are the following:

 SYMM x,y,z                     Atom: Mn_1      0.5000  0.0000  0.0000
 Sk(1): (u,v,w)
 SYMM -x+1,-y,z+1/2             Atom: Mn_2      0.5000  0.0000  0.5000
 Sk(2): (-u,-v,w)
 SYMM -x+1/2,y+1/2,-z+1/2       Atom: Mn_3      0.0000  0.5000  0.5000
 Sk(3): (u,-v,w)
 SYMM x-1/2,-y+1/2,-z           Atom: Mn_4      0.0000  0.5000  0.0000
 Sk(3): (-u,v,w)
```

The fact that only four *irreps* contribute to the magnetic representation is due to the fact that the Wyckoff position (4b) contains a centre of symmetry and the *irreps* with $\chi(\bar{1})$=-1 are not allowed. In other terms, time inversion cannot be mixed with the centre of symmetry in this

case because the atoms in that position would have a zero magnetic moment. The point group $\bar{1}'$ is not admissible (see section 2.3) .

The interpretation of the above table in terms of the mathematical expressions given in this article is as follows (the indices $n\lambda$ are combined in a single integer index):

Example 1: the atomic component of the basis vector (*BsV*) 1 for atom Mn_2 of representation Γ_3 for propagation vector (000) is: $\mathbf{S}_{n\lambda}^{\mathbf{k}\nu}(js) = \mathbf{S}_1^{(000)\,3}\ Mn_2\ = (-1, 0, 0)$

Example 2: The atomic component of the basis vector 2 for atom Mn_3 of representation Γ_3 for propagation vector (000) is: $\mathbf{S}_{n\lambda}^{\mathbf{k}\nu}(js) = \mathbf{S}_2^{(000)\,3}\ Mn_3\ = (0, -1, 0)$

Example 3: the full basis vector 2 for representation Γ_3 and propagation vector (000) for the whole set of atoms is:

$$BsV(2,1:4) \rightarrow \boldsymbol{\psi}_{n\lambda}^{\mathbf{k}\nu} = \boldsymbol{\psi}_2^{(000)\,3} = (\overset{1}{\overbrace{0,\ 1,\ 0}},\ \overset{2}{\overbrace{0,-1,\ 0}},\ \overset{3}{\overbrace{0,-1,\ 0}},\ \overset{4}{\overbrace{0,\ 1,\ 0}})$$

If we call u, v, w the three free mixing coefficients (in our case they are real numbers because $\mathbf{k}=0$), the magnetic structure can be globally described by the global Fourier coefficient (it coincides with the whole set of magnetic moments):

$$\mathbf{m}_{[1,2,3,4]} = \mathbf{S}_{[\mathbf{k}]} = \sum_{n\lambda} C_{n\lambda}^{\mathbf{k}\nu}\boldsymbol{\psi}_{n\lambda}^{\mathbf{k}\nu} = u\boldsymbol{\psi}_1^{\mathbf{k}\nu} + v\boldsymbol{\psi}_2^{\mathbf{k}\nu} + w\boldsymbol{\psi}_3^{\mathbf{k}\nu}$$

Figure 5: Magnetic structure of LaMnO$_3$. Four unit cells and the numbering of the Mn atoms are shown. From the fitting of the powder diffraction pattern, we obtain $u \approx 0$, $v \approx 3.8\mu_B$, $w \approx 0$. See text for details.
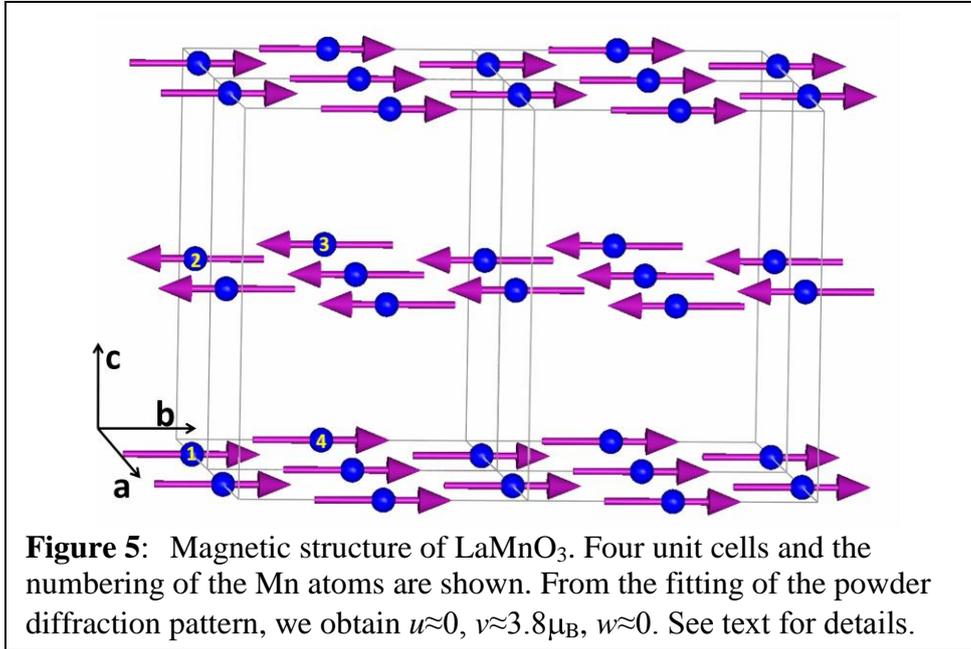
The individual magnetic moments of the four atoms are:

$$Sk(1) \rightarrow \mathbf{m}_1 = \mathbf{S}_{\mathbf{k}1} = \sum_{n\lambda} C_{n\lambda}^{\mathbf{k}\nu}\mathbf{S}_{n\lambda}^{\mathbf{k}\nu}(1) = (u, v, w); \qquad Sk(2) \rightarrow \mathbf{m}_2 = \mathbf{S}_{\mathbf{k}2} = \sum_{n\lambda} C_{n\lambda}^{\mathbf{k}\nu}\mathbf{S}_{n\lambda}^{\mathbf{k}\nu}(2) = (-u, -v, w)$$

$$Sk(3) \rightarrow \mathbf{m}_3 = \mathbf{S}_{\mathbf{k}3} = \sum_{n\lambda} C_{n\lambda}^{\mathbf{k}\nu}\mathbf{S}_{n\lambda}^{\mathbf{k}\nu}(3) = (u, -v, w); \quad Sk(4) \rightarrow \mathbf{m}_4 = \mathbf{S}_{\mathbf{k}4} = \sum_{n\lambda} C_{n\lambda}^{\mathbf{k}\nu}\mathbf{S}_{n\lambda}^{\mathbf{k}\nu}(4) = (-u, v, w)$$

A very common notation in the literature is that of sequence of signs G(+,−,+,−), A(+,−,−,+), F(+,+,+,+) and C(+,+,−,−), called *modes* by Bertaut [6,7]. For the current *irrep* Γ_3 (Shubnikov group *Pb'n'm*) the label for the magnetic structure in terms of these modes is: (G_x, A_y, F_z). The structure is antiferromagnetic with a very weak ferromagnetic component (only seen by macroscopic magnetisation measurements) along **c** and formed by ferromagnetic planes

stacked antiferromagnetically along **c**. This is the so called A-type AF structure in literature about perovskites. The structure is shown in Figure 5.

If we list the four magnetic moments (identical to Fourier coefficients in this case) as:

$1(u,v,w)$; $2(-u,-v,w)$; $3(u,-v,w)$; $4(-u,v,w)$

together with the symmetry operators that pass from atom 1 to 1, 2, 3, 4, respectively:

$1(x,y,z):1$; $2(-x+1,-y,z+1/2): 2_{1z}$; $3(-x+1/2, y+1/2,-z+1/2): 2_{1y}$; $4(x-1/2,-y+1/2,-z): 2_{1x}$

we can see that the rotational parts of the symmetry operators correspond to the action of the elements: 1, 2_z, 2_y and 2_x respectively. We can interpret the symbols (u, v, w) as matrices corresponding to the transformation of the magnetic moment of the atom 1 to the magnetic moments of the atoms 1,2,3,4. As binary axes are proper rotations, we can see that the matrices correspond to the symmetry operators: 1, 2_z, $2'_y$ and $2'_x$ respectively. Time inversion is then associated with the symmetry operators 2_{1y} and 2_{1x} as required by the Shubnikov group *Pb'n'm* (see the characters of the *irrep* Γ_3 for operators $\{2_x|½½0\}$ and $\{2_y|½½½\}$ in Table 1).

10.2: *Symmetry analysis of the magnetic structure of* KTb_3F_{12}.

This compound contains two Tb sites, 2*b* and 4*d*, which are attributed to a charge ordering of Tb^{3+} and Tb^{4+} respectively [27]. Additional information can be found in the "Examples and Tutorials" section of the FullProf site [28]. The space group is **G**=*I4/m*, the cell parameters are a=7.695 Å and c= 7.540 Å at room temperature. The first magnetic peak can be indexed as (100) that is a forbidden reflection for a lattice of *I*-type. That means the propagation vector **k** = (1, 0, 0) = ½ (2, 0, 0) is in the surface of the Brillouin Zone and it is equivalent to –**k**. The group of the propagation vector is the whole space group because all symmetry operators leave invariant the propagation vector: **G$_k$** = **G** = *I4/m*. The irreducible representations are given in Table 2.

Table 2: Irreducible representations of **G$_k$**=**G**= *I4/m* for **k**=(1, 0, 0)

Irreps	{1}	{2$_z$}	{4^+_z}	{4^-_z}	{-1}	{m$_z$}	{-4^+_z}	{-4^-_z}	SG
Γ_1:	1	1	1	1	1	1	1	1	I_p4/m
Γ_2:	1	1	1	1	-1	-1	-1	-1	I_p4/m'
Γ_3:	1	1	-1	-1	1	1	-1	-1	I_p4'/m
Γ_4:	1	1	-1	-1	-1	-1	1	1	I_p4'/m'
Γ_5:	1	-1	i	-i	1	-1	i	-i	
Γ_6:	1	-1	i	-i	-1	1	-i	i	
Γ_7:	1	-1	-i	i	1	-1	-i	i	
Γ_8:	1	-1	-i	i	-1	1	i	-i	

Here the small representations coincide with the *irreps* of the full space group. The complex *irreps* Γ_5 to Γ_8 are of dimension 1, so they cannot be reduced to real representations because matrices of dimension 1 always commute: there is no similarity unitary transformation reducing them to real matrices. This is the reason there is no Shubnikov symbol associated to them. Notice that Γ_5 is the complex conjugate of Γ_7 and Γ_6 is the complex conjugate of Γ_8. We are in the case 2 of rule (69). Let us consider now the position of the magnetic atoms Tb(2*b*): (0,0,1/2) and Tb(4*d*): (0,1/2,1/4), (1/2,0,1/4). Notice that we consider only the content

of a primitive cell even if we refer the coordinates to the conventional centred cell. The global magnetic representation obtained by the direct product of the axial representation by the permutation representation has the following list of characters for the symmetry operators of the space group given in the same order as in Table 2:

$$\chi\,(\Gamma_m\text{-}2b) = (3,-1,\ 1,\ 1,\ 3,-1,\ 1,\ 1)$$
$$\chi\,(\Gamma_m\text{-}4d) = (6,-2,\ 0,\ 0,\ 0,\ 0,-2,-2)$$

The decomposition of the global magnetic representation in terms of *irreps* for the two sites is given by:

$$\Gamma_m(2b) = \Gamma_1 \oplus \Gamma_5 \oplus \Gamma_7$$

$$\Gamma_m(4d) = \Gamma_2 \oplus \Gamma_3 \oplus \Gamma_5 \oplus \Gamma_6 \oplus \Gamma_7 \oplus \Gamma_8$$

The final Fourier coefficients obtained from the basis functions calculated for the different representations and the two sites are summarised in the following:

$$\Gamma_1(2b):\mathbf{S_k} = (0,0,v)$$

Site 2*b*, a single sublattice:
$$\Gamma_5(2b):\mathbf{S_k} = (v,0,0) + i(0,-v,0)$$
$$\Gamma_7(2b):\mathbf{S_k} = (v,0,0) + i(0,\ \ v,0)$$

Site 4*d*, two sublattices:
$$\Gamma_2(4d):\mathbf{S_k}(1) = (0,0,u); \qquad \mathbf{S_k}(2) = (0,0,-u)$$
$$\Gamma_3(4d):\mathbf{S_k}(1) = (0,0,u); \qquad \mathbf{S_k}(2) = (0,0,u)$$
$$\Gamma_5(4d):\mathbf{S_k}(1) = (u,0,0) + i(0,u,0); \quad \mathbf{S_k}(2) = (u,0,0) + i(0,u,0)$$
$$\Gamma_6(4d):\mathbf{S_k}(1) = (u,0,0) + i(0,-u,0); \quad \mathbf{S_k}(2) = (-u,0,0) + i(0,u,0)$$
$$\Gamma_7(4d):\mathbf{S_k}(1) = (u,0,0) + i(0,-u,0); \quad \mathbf{S_k}(2) = (u,0,0) + i(0,-u,0)$$
$$\Gamma_8(4d):\mathbf{S_k}(1) = (u,0,0) + i(0,u,0); \quad \mathbf{S_k}(2) = (-u,0,0) + i(0,-u,0)$$

The parameters u and v correspond to the coefficients $C_{n\lambda}^{\mathbf{k}\nu}$ and are the free parameters of the possible magnetic structures. Here we have simplified the notation but one has to keep in mind that the parameters appearing in different representations may have different values. The representations Γ_1, Γ_2 and Γ_3 describe real magnetic structures for this system. Notice that the representation Γ_4 does not appear in the decomposition of the global magnetic representation for any of the two sites. One can see that the *irreps* Γ_5, Γ_6, Γ_7 and Γ_8 cannot describe alone real magnetic structures because, according to the eq. (34), the Fourier coefficients must be real. Only if we combine the basis functions of the *irreps* that are complex conjugate, and we put the same value for the coefficients, we can obtain real magnetic structures. So, apart from the three representations mentioned above the following combination or representations and basis functions give real magnetic structures:

$$\Gamma_1,\quad \Gamma_2,\quad \Gamma_3,\quad \Gamma_5 \oplus \Gamma_7,\quad \Gamma_6 \oplus \Gamma_8$$

The corresponding basis functions are the following:

Site 2*b*, a single sublattice:
$$\Gamma_1(2b):\mathbf{S_k} = (0,0,v)$$
$$\Gamma_5 \oplus \Gamma_7(2b):\mathbf{S_k} = (v,0,0)$$

$$\Gamma_2(4d):\mathbf{S_k}(1)=(0,0,u);\quad \mathbf{S_k}(2)=(0,0,-u)$$
$$\Gamma_3(4d):\mathbf{S_k}(1)=(0,0,u);\quad \mathbf{S_k}(2)=(0,0,u)$$

Site 4d, two sublattices:
$$\Gamma_5\oplus\Gamma_7(4d):\mathbf{S_k}(1)=(u,0,0);\quad \mathbf{S_k}(2)=(u,0,0)$$
$$\Gamma_6\oplus\Gamma_8(4d):\mathbf{S_k}(1)=(u,0,0);\quad \mathbf{S_k}(2)=(-u,0,0)$$

When more than one site exists in a crystallographic structure, the magnetic ordering is usually driven by a single *irrep*. So, in our case if magnetic ordering should exist in both sites the only common magnetic structure should be described within the reducible representation $\Gamma_5\oplus\Gamma_7$ (called usually *physically irreducible*).
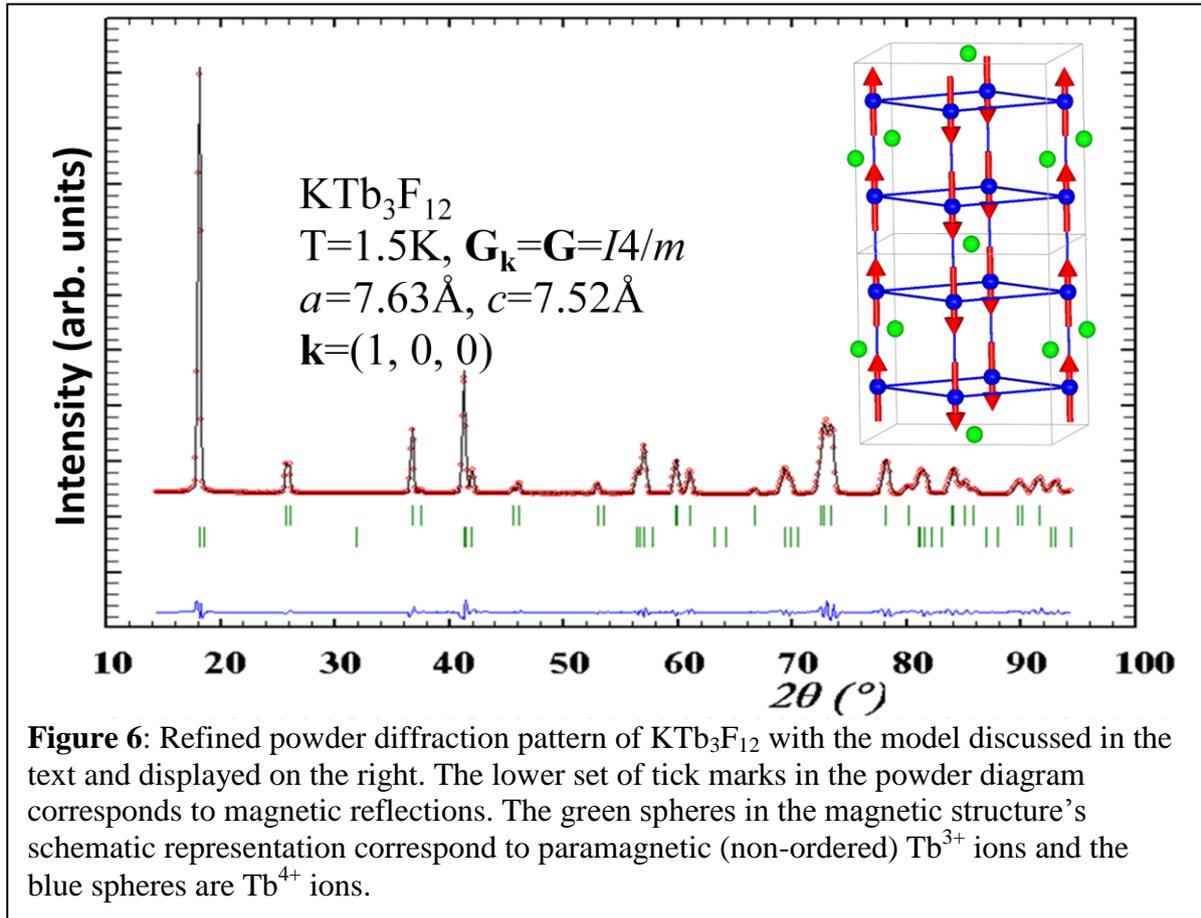
Figure 6: Refined powder diffraction pattern of KTb_3F_{12} with the model discussed in the text and displayed on the right. The lower set of tick marks in the powder diagram corresponds to magnetic reflections. The green spheres in the magnetic structure's schematic representation correspond to paramagnetic (non-ordered) Tb^{3+} ions and the blue spheres are Tb^{4+} ions.

Experimentally, the *physically irreducible* representation $\Gamma_5\oplus\Gamma_7$ does not give satisfactory results. The analysis of the diffraction pattern shows that the site 4d (Tb^{4+}) orders according to the single *irrep* Γ_3, which does not exist for site 2b, so the magnetic moment of atoms in 2b (Tb^{3+}) should be zero. This is easily verified experimentally and well justified from the physical point of view because the crystal field acting on a Tb^{3+} may create a non magnetic singlet. Iin this case, the molecular field of the magnetic configuration of Tb^{4+} ions creates a zero molecular field at the site of Tb^{3+} so that, even in the case of an effective magnetic doublet, the Tb^{3+} ions remain paramagnetic due to frustration. The refined powder diffraction pattern at 1.5K and a scheme of the magnetic structure are shown in Figure 6.

10.3: *Symmetry analysis of the magnetic structure of the compounds* R_2BaMiO_5.

The magnetic structures of the family of compounds R_2BaNiO_5 (R = Rare earth or Y) have been studied and summarised in reference [28]. The R_2BaNiO_5 oxides present interesting structural and magnetic properties due to the fact that their structure possesses a strong 1D character [4, 5]. These oxides crystallise in the orthorhombic system with the body centred space group $\mathbf{G}$ = *Immm* and having approximate cell parameters: $a \approx 3.8$ Å, $b \approx 5.8$ Å, and $c \approx 11.3$ Å. The main structural feature is the presence of one-dimensional (1D) chains of NiO_6 octahedra along the *a*-axis. The octahedra are strongly distorted with a very short $Ni-O_{apical}$ distance (≈ 1.88 Å), and a longer $Ni-O_{basal}$ distance (≈ 2.18 Å). The basal angle O-Ni-O ($\approx 78°$) is also much smaller than expected for a regular octahedron ($90°$). The structural details at room temperature and the dependence of the rare earth size upon the structural parameters have been extensively discussed in reference [29].

The Ni ions are in a single Bravais sublattice at (2*a*) site: (0, 0, 0). The rare earth site (4*j*) generates two sublattices 1: (1/2, 0, *z*) and 2: (−1/2, 0, −*z*) in a primitive unit cell. The other atoms of the conventional unit cell are related to those already given by the centring translation $\mathbf{t}_I$ =(1/2, 1/2, 1/2). The magnetic moments in positions outside the reference cell are calculated from those at the reference cell by the expression (34). Thus, in order to describe the magnetic structure we need to know only the Fourier components of magnetic moments of the Ni ion at the origin and of R_1 at (1/2, 0, *z*) and R_2 at (−1/2, 0, −*z*).

These compounds are all antiferromagnetic with propagation vector $\mathbf{k}$=(1/2, 0, 1/2), equivalent to -$\mathbf{k}$. However the directions of magnetic moments are different as a consequence of the different anisotropy of the rare earth ions. The symmetry analysis using the representation theory and the program BASIREPS [24], provides immediately the little co-group $\mathbf{G_{0k}}$=2/*m*, with unique axis along $\mathbf{b}$. The representations of $\mathbf{G_k}$ are those of $\mathbf{G_{0k}}$ because the space group is symmorphic, there are then four 1D *irreps* of $\mathbf{G_k}$. The small representations are gathered in Table 3.

Table 3: Small irreducible representations of $\mathbf{G_k}$= *I*12/*m*1 for $\mathbf{k}$=(1/2, 0, 1/2) in $\mathbf{G}$=*Immm*.

irreps	{1|000}	{2$_y$|000}	{-1|000}	{m$_y$|000}
Γ_1 (A$_g$):	1	1	1	1
Γ_2 (A$_u$):	1	1	-1	-1
Γ_3 (B$_g$):	1	-1	1	-1
Γ_4 (B$_u$):	1	-1	-1	1

We have added the notation for the *irreps* used in reference [28] and in chapters by Perez-Mato *et al.* and Tasci *et al.* The star of the $\mathbf{k}$-vector is constituted by two independent arms:

$$\{\mathbf{k}\} = \{\mathbf{k}=\mathbf{k}_1=(1/2,\ 0,\ 1/2);\ \mathbf{k}_2=(-1/2,\ 0,\ 1/2)\}$$

The space group can be decomposed in cosets (eq. (27)) as: $\mathbf{G} = \mathbf{G_k} + m_x \mathbf{G_k}$, where m_x is the mirror plane perpendicular to the $\mathbf{a}$-axis that transforms $\mathbf{k}_1$ into $\mathbf{k}_2$.

The full representations of the space group *Immm* are two dimensional and can be constructed by using the induction formula (30). As an example of the use of formula (30) we give below the 2D matrix corresponding to the operator 2_y for *irrep* Γ_3:

$$\Gamma_{1,1}^{\{\mathbf{k}\}3}(2_y) = \Gamma^{\mathbf{k}3}(g_1^{-1}2_y\,g_1)\delta_{g_1^{-1}2_y\,g_1\in G_\mathbf{k}} = \Gamma^{\mathbf{k}3}(1\,2_y\,1)\delta_{12_y\,1\in G_\mathbf{k}} = \Gamma^{\mathbf{k}3}(2_y)\delta_{2_y\in G_\mathbf{k}} = -1$$

$$\Gamma_{2,2}^{\{\mathbf{k}\}3}(2_y) = \Gamma^{\mathbf{k}3}(g_2^{-1}2_y\,g_2)\delta_{g_2^{-1}2_y\,g_2\in G_\mathbf{k}} = \Gamma^{\mathbf{k}3}(m_x\,2_y\,m_x)\delta_{m_x2_y\,m_x\in G_\mathbf{k}} = \Gamma^{\mathbf{k}3}(2_y)\delta_{2_y\in G_\mathbf{k}} = -1$$

$$\Gamma_{1,2}^{\{\mathbf{k}\}3}(2_y) = \Gamma^{\mathbf{k}3}(g_1^{-1}2_y\,g_2)\delta_{g_1^{-1}2_y\,g_2\in G_\mathbf{k}} = \Gamma^{\mathbf{k}3}(1\,2_y\,m_x)\delta_{12_y\,m_x\in G_\mathbf{k}} = \Gamma^{\mathbf{k}3}(m_z)\delta_{m_z\in G_\mathbf{k}} = 0$$

$$\Gamma_{2,1}^{\{\mathbf{k}\}3}(2_y) = \Gamma^{\mathbf{k}3}(g_2^{-1}2_y\,g_1)\delta_{g_1^{-1}2_y\,g_2\in G_\mathbf{k}} = \Gamma^{\mathbf{k}3}(m_x2_y\,1)\delta_{m_x2_y\,1\in G_\mathbf{k}} = \Gamma^{\mathbf{k}3}(m_z)\delta_{m_z\in G_\mathbf{k}} = 0$$

$$\Gamma^{\{\mathbf{k}\}3}(2_y) = \begin{pmatrix} -1 & 0 \\ 0 & -1 \end{pmatrix}$$

The matrix corresponding to m_z that does not belong to $\mathbf{G_k}$ is given by:

$$\Gamma_{1,1}^{\{\mathbf{k}\}3}(m_z) = \Gamma^{\mathbf{k}3}(g_1^{-1}m_z g_1)\delta_{g_1^{-1}m_z\,g_1\in G_\mathbf{k}} = \Gamma^{\mathbf{k}3}(1\,m_z\,1)\delta_{1m_z\,1\in G_\mathbf{k}} = \Gamma^{\mathbf{k}3}(m_z)\delta_{m_z\in G_\mathbf{k}} = 0$$

$$\Gamma_{2,2}^{\{\mathbf{k}\}3}(m_z) = \Gamma^{\mathbf{k}3}(g_2^{-1}m_z\,g_2)\delta_{g_2^{-1}m_z\,g_2\in G_\mathbf{k}} = \Gamma^{\mathbf{k}3}(m_x\,m_z\,m_x)\delta_{m_x m_z\,m_x\in G_\mathbf{k}} = \Gamma^{\mathbf{k}3}(m_z)\delta_{m_z\in G_\mathbf{k}} = 0$$

$$\Gamma_{1,2}^{\{\mathbf{k}\}3}(m_z) = \Gamma^{\mathbf{k}3}(g_1^{-1}m_z\,g_2)\delta_{g_1^{-1}m_z\,g_2\in G_\mathbf{k}} = \Gamma^{\mathbf{k}3}(1\,m_z\,m_x)\delta_{1m_z\,m_x\in G_\mathbf{k}} = \Gamma^{\mathbf{k}3}(2_y)\delta_{2_y\in G_\mathbf{k}} = -1$$

$$\Gamma_{2,1}^{\{\mathbf{k}\}3}(m_z) = \Gamma^{\mathbf{k}3}(g_2^{-1}m_z g_1)\delta_{g_1^{-1}m_z\,g_2\in G_\mathbf{k}} = \Gamma^{\mathbf{k}3}(m_x m_z\,1)\delta_{m_x m_z\,1\in G_\mathbf{k}} = \Gamma^{\mathbf{k}3}(2_y)\delta_{2_y\in G_\mathbf{k}} = -1$$

$$\Gamma^{\{\mathbf{k}\}3}(m_z) = \begin{pmatrix} 0 & -1 \\ -1 & 0 \end{pmatrix}$$

One can proceed similarly for all the coset representatives of the space group *Immm* and we obtain all the irreducible representations of *Immm* (from all the small *irreps* of $\mathbf{G_k}$) that are all two dimensional for the star $\{\mathbf{k}_1, \mathbf{k}_2\}$. Normally we do not need to calculate explicitly these matrices because the basis vectors corresponding to the different arms are given explicitly by the formula (55).

Let us come back to the small representations for the single $\mathbf{k} = (1/2, 0, 1/2)$. The global magnetic reducible representation has the following list of characters for the symmetry operators of the space group given in the same order as in the above table (1, 2_y, -1, m_y):

$$\chi\,(\Gamma_m\text{-Ni}) = (3, -1,\ 3,\ -1)$$
$$\chi\,(\Gamma_m\text{-R}) = (6,\ 0,\ 0,\ -2)$$

The decomposition of the global magnetic representation in terms of *irreps* for the two sites is given by:

$$\Gamma_m(Ni) = \Gamma_1 \oplus 2\Gamma_3$$

$$\Gamma_m(R) = \Gamma_1 \oplus 2\Gamma_2 \oplus 2\Gamma_3 \oplus \Gamma_4$$

In principle (if the two sites order according to the same representation) only the representations Γ_1 and Γ_3 are allowed. The final Fourier coefficients obtained from the basis functions calculated for these representations and the two sites are summarised in the following:

$$\Gamma_1: \quad \text{Ni}\,(0, v, 0) \qquad R_1(0, q, 0),\ R_2(0, q, 0)$$
$$\Gamma_3: \quad \text{Ni}\,(u, 0, w) \qquad R_1(p, 0, r),\ R_2(p, 0, r)$$

The coefficients should be real because **k** is equivalent to –**k**. The experiments have demonstrated that the active representation is Γ_3 (four free parameters: u, w, p, r) for all the rare earth except for Tm in which the data can only be explained by mixing both Γ_1 and Γ_3 (six free parameters) with a strongly dominant Γ_1 component (within 3 standard deviations in the magnetic moments, only Γ_1 is present in the Tm case). Notice that (see Table 3), from the point of view of symmetry invariance, the representation Γ_3 corresponds to having the following association of symmetry operators of the little co-group with time inversion: {1, $2'_y$, -1, m'$_y$}. Just the trivial {1, 2_y, -1, m_y} colourless little co-group is associated with Γ_1.

As an example we show the refinement of the powder diffraction pattern of Ho_2BaNiO_5 and a picture of its magnetic structure, assumed to be single **k**.

Figure 7: Refined powder diffraction pattern of Ho_2BaNiO_5 and scheme of its magnetic structure. The space group is *Immm*. There is only three magnetic atoms per primitive unit cell. Ni at (0, 0, 0) and Ho at positions 1(1/2, 0, z), 2(-1/2, 0,-z) with z=0.2025. The magnetic moments are not in scale, the amplitude of those of Ho atoms have been arbitrarily multiplied by 0.3 for representation purposes. The magnetic unit cell is doubled along **a** and **c**. The magnetic moments of the three atoms are: Ni(u, 0, w), with $u \approx 0.59\mu_B$, $w \approx -1.3\mu_B$, Ho$_1$(p, 0, r), Ho$_2$(p, 0, r), with $p \approx 0.1\mu_B$ and $r \approx 9\mu_B$.

With powder diffraction it is, in general, impossible to determine the transition channel (e.g. what are the arms of the star defining the magnetic structure), however if we assume that the magnetic structure is multi-**k** in this case we will show that the magnetic moments amplitude will not be constant. Let us just select the Ni ions at the origin of the lattice. The Fourier sum (34) can be written for the present case as:

$$\mathbf{m}_l = \mathbf{S}_{\mathbf{k}_1} exp(-\pi i(l_1 + l_3)) + \mathbf{S}_{\mathbf{k}_2} exp(-\pi i(-l_1 + l_3))$$

$$\mathbf{m}_l = \begin{cases} \pm(\mathbf{S}_{\mathbf{k}_1} + \mathbf{S}_{\mathbf{k}_2}); & l_1, l_3 \in \mathbb{Z} \\ \pm(\mathbf{S}_{\mathbf{k}_1} - \mathbf{S}_{\mathbf{k}_2}); & l_1 = \frac{1}{2} + n_1; \ l_3 = \frac{1}{2} + n_3; \quad n_1, n_3 \in \mathbb{Z} \end{cases}$$

The real Fourier coefficients $\mathbf{S}_{k1}$ and $\mathbf{S}_{k2}$ may be chosen in order to restore the global orthorhombic symmetry imposing that m_x, or its combination with time inversion, has to be a

symmetry operator of the magnetic structure. In any case one can see that the magnetic moment amplitude can only be conserved if S_{k1} is perpendicular to S_{k2}. If one applies the formula (55) taking into account (56), for the basis vectors using $h_L=m_x$ (that transform k_1 into k_2) the basis vectors (except for a change of sign) are the same for Ni ions. This implies that the Fourier coefficients S_{k1} and S_{k2} are parallel, so the amplitude of the magnetic moments cannot be conserved in a multi-k structure for the present case.

10.4: *Symmetry analysis of the commensurate magnetic structures of the compounds* R_2T_2X.

The R_2T_2X compounds, with R = Rare-Earth or Uranium, T = Fe, Co, Ni, Ru, Rh or Pd and X= Sn or In, crystallise in the tetragonal U_3Si_2-type crystal structure with space group *P4/mbm* [30-35]. The occupied Wyckoff positions are (4*h*) R: (x_R, 1/2+x_R, 1/2); (4*g*) T: (x_T, 1/2+x_T,0) and X: (2*a*) (0, 0, 0). The R-ions are the only "magnetic" atoms all along the R_2T_2X series of compounds.

Some examples of commensurate magnetic structures, revealed from neutron diffraction experiments in this family of compounds, are shown in Figure 8. These are described either with k = (0 0 0), identical crystalline and magnetic unit-cells, or k = (0 0 ½) (*a*, *a*, 2*c* magnetic unit-cell). Other commensurate k = (½ ½ 0), k = (½ ½ ½) and k = (¼ ¼ ½) values have been observed for compounds in the series, but with no example shown in Figure 8.

8a
$U_2Pd_2In - U_2Pd_2Sn - k = (0\ 0\ 0)$

8b
$U_2Pd_{2.4}Sn_{0.6} - k = (0\ 0\ ½)$

8c
$U_2Ni_2In - k = (0\ 0\ ½)$

8d
$U_2Ni_2Sn - U_2Rh_2Sn - k = (0\ 0\ ½)$
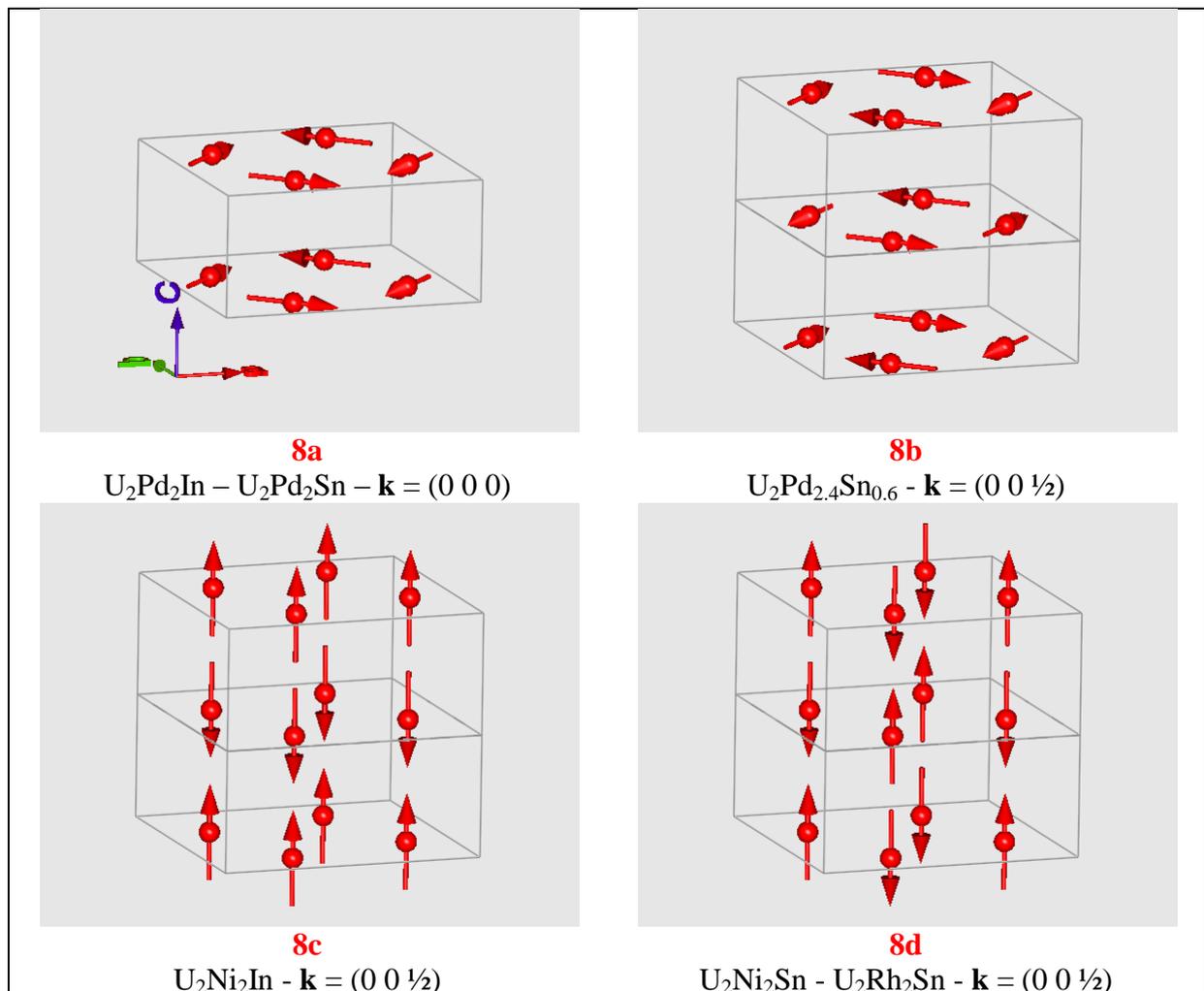
Figure 8: Commensurate magnetic structures in *P4/mbm* R_2T_2X compounds [30-34].

We shall take as examples the symmetry analysis for the cases **k** = (0, 0, 0) and **k** = (0, 0, 1/2) corresponding to the compounds shown in Figure 8. The determination of their magnetic structure has been performed using the method that we have illustrated in the previous examples. Here we will give some more detailed information concerning the reducible *magnetic representation* discussed in sections 7.1 and 7.2.

Table 4 lists the symmetry operators of the coset representatives of *P4/mbm* space group with respect to the translation group. When no translation associated to the symmetry operator is given, [000] is assumed. The same applies for the location of the symmetry element. We have emphasised the lines containing the generators in the Hermann-Mauguin (H-M) symbol, and written them in red.

Table 4: Coset representatives of the space group **G**= *P4/mbm*

Symmetry operators ordering	Symmetry operators	Symbol (rotational part)	Translation	Location	Kovalev Notation (rotational part)
SYMM(1)	*x, y, z*	1			h_1
SYMM(2)	*-x, -y, z*	2_z			h_4
SYMM(3)	*½-x, ½+y, -z*	2_y	[0 ½ 0]	(¼ 0 0)	h_3
SYMM(4)	*½+x, ½-y, -z*	2_x	[½ 0 0]	(0 ¼ 0)	h_2
SYMM(5)	*½+y, ½+x, -z*	$2_{[110]^-}$	[½ ½ 0]		h_{16}
SYMM(6)	*½-y, ½-x, -z*	$2_{[110]^-}$		(0 ½ 0)	h_{13}
SYMM(7)	*y, -x, z*	4_z^3			h_{15}
SYMM(8)	*-y, x, z*	4_z (4)			h_{14}
SYMM(9)	*-x, -y, -z*	$\bar{1}$			h_{25}
SYMM(10)	*x, y, -z*	m_z (/m)			h_{28}
SYMM(11)	*½+x, ½-y, z*	m_y	[½ 0 0]	(0 ¼ 0)	h_{27}
SYMM(12)	*½-x, ½+y, z*	m_x (b)	[0 ½ 0]	(¼ 0 0)	h_{26}
SYMM(13)	*½-y, ½-x, z*	$m_{[110]^-}$(m)		(0 ½ 0)	h_{40}
SYMM(14)	*½+y, ½+x, z*	$m_{[110]^-}$	[½ ½ 0]		h_{37}
SYMM(15)	*-y, x, -z*	$\bar{4}_z^3$			h_{39}
SYMM(16)	*y, -x, -z*	$\bar{4}_z$			h_{38}

 The U atoms (the only magnetic atoms) are labelled from 1 to 4, as shown below, either in the table or in the figure. The corresponding Fourier coefficients (in this case identical to magnetic moments) are referred as S_1, S_2, S_3 and S_4.

Table 5: List of U-atoms in the 4h positions of space group **G** = *P4/mbm*. The list of symmetry operators passing from the first atom to the others is given as well as the coordinates in terms of the parameter x_U.

		SYMM order + translation	x	y	z
U_1	x, y, z	(1)	x_U	$\frac{1}{2} + x_U$	$\frac{1}{2}$
U_2	$-x+1, -y+1, z$	(2)+[1,1,0]	$1 - x_U$	$\frac{1}{2} - x_U$	$\frac{1}{2}$
U_3	$-x+1/2, y-1/2, -z+1$	(3)+[0,-1,1]	$\frac{1}{2} - x_U$	x_U	$\frac{1}{2}$
U_4	$x+1/2, -y+3/2, -z+1$	(4)+[0,1,1]	$\frac{1}{2} + x_U$	$1 - x_U$	$\frac{1}{2}$
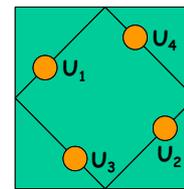
In the 12-dimensional space of S_1 to S_4 magnetic moments components (12=4×3, where 4 stands for the number of distinct atoms, and 3 for the number of magnetic moment components for one atom), each of the G_k symmetry operators is represented by a 12×12 square matrix: this matrix reflects the transformations of S_1, S_2, S_3 and S_4 by the considered symmetry operator, including both permutational and orientational contributions (permutation of U_{1-4} atoms, (re)orientation of magnetic moments). The components of the matrices can be obtained simply by inspection of the action of the symmetry operator (taking into account the axial character of **S**-vectors) or by applying directly the formula (49) or (50) and (51).

For instance, the magnetic matrix for SYMM(1) is clearly the 12×12 identity matrix with character $\chi_m(1)=12$. For displaying the magnetic representation matrices we can use a cell representation of the matrices in order to emphasize the labelling of the atoms positions. The magnetic matrices for the SYMM(10) operator (mirror perpendicular to the tetragonal **c**-axis, keeping unchanged U_{1-4} atoms) and for SYMM(2) operator (two-fold axis parallel to the **c**-axis, that permutes atoms 1-2 and 3-4) are:

$\Gamma_m(m_z)=\Gamma_m(10)=$

	U_1	U_2	U_3	U_4
U_1	-1 -1 1			
U_2		-1 -1 1		
U_3			-1 -1 1	
U_4				-1 -1 1

of character $\chi_m(10)=-4$, and

$\Gamma_m(2_z)=\Gamma_m(2)=$

	U_1	U_2	U_3	U_4
U_1		-1 -1 **1**		
U_2	-1 -1 1			
U_3				-1 -1 1
U_4			-1 -1 1	

of character $\chi_m(2)=0$. Each cell is a 3 x 3 matrix holding the axial representation of the operator. Empty cells are filled with zeroes in the conventional representation of a matrix.

The set of all the sixteen magnetic matrices form a 12-dimensional representation (Γ_m) of the $\mathbf{G_{k=(000)}}$ group. The matrices for $\mathbf{k}=(0, 0, 1/2)$ are constructed in a similar way and the differences come from the phase factors we have to include when using "returning vectors" that in our case are always +1 or -1. The explicit form of the matrices can easily be constructed by inspecting the output of the program BASIREPS [24].

Γ_m can be decomposed into *irreps* of $\mathbf{G_{k=(000)}}$ or $\mathbf{G_{k=(001/2)}}$. These *irreps* coincide in this particular case (notice that there is no symmetry operator in the list of the previous table with associated translation along the **c**-axis) and are listed in Table 6. The numbers of the first row correspond to the symmetry operators listed in a previous table with operators 8, 10, 12 and 13 emphasised because they appear in the H-M symbol of the space group.

Table 6: Irreducible representations of $\mathbf{G_k}= \mathbf{G}=P4/mbm$ for $\mathbf{k}=(0, 0, 0)$ and $\mathbf{k}=(0, 0, 1/2)$.

P4/mbm k=(0,0,0) k=(0,0,1/2)	1	2	3	4	5	6	7	8 (4)	9	10 (/m)	11	12 (b)	13 (m)	14	15	16
Γ_1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1
Γ_2	1	1	1	1	1	1	1	1	-1	-1	-1	-1	-1	-1	-1	-1
Γ_3	1	1	1	1	-1	-1	-1	-1	1	1	1	1	-1	-1	-1	-1
Γ_4	1	1	1	1	-1	-1	-1	-1	-1	-1	-1	-1	1	1	1	1
Γ_5	1	1	-1	-1	1	1	-1	-1	1	1	-1	-1	1	1	-1	-1
Γ_6	1	1	-1	-1	1	1	-1	-1	-1	-1	1	1	-1	-1	1	1
Γ_7	1	1	-1	-1	-1	-1	1	1	1	1	-1	-1	-1	-1	1	1
Γ_8	1	1	-1	-1	-1	-1	1	1	-1	-1	1	1	1	1	-1	-1
Γ_9	$\begin{smallmatrix}1&0\\0&1\end{smallmatrix}$	$\begin{smallmatrix}-1&0\\0&-1\end{smallmatrix}$	$\begin{smallmatrix}1&0\\0&-1\end{smallmatrix}$	$\begin{smallmatrix}-1&0\\0&1\end{smallmatrix}$	$\begin{smallmatrix}0&1\\1&0\end{smallmatrix}$	$\begin{smallmatrix}0&-1\\-1&0\end{smallmatrix}$	$\begin{smallmatrix}0&-1\\1&0\end{smallmatrix}$	$\begin{smallmatrix}0&1\\-1&0\end{smallmatrix}$	$\begin{smallmatrix}-1&0\\0&-1\end{smallmatrix}$	$\begin{smallmatrix}1&0\\0&1\end{smallmatrix}$	$\begin{smallmatrix}-1&0\\0&1\end{smallmatrix}$	$\begin{smallmatrix}1&0\\0&-1\end{smallmatrix}$	$\begin{smallmatrix}0&-1\\0&-1\end{smallmatrix}$	$\begin{smallmatrix}0&1\\1&0\end{smallmatrix}$	$\begin{smallmatrix}0&1\\-1&0\end{smallmatrix}$	$\begin{smallmatrix}0&-1\\1&0\end{smallmatrix}$
Γ_{10}	$\begin{smallmatrix}1&0\\0&1\end{smallmatrix}$	$\begin{smallmatrix}-1&0\\0&-1\end{smallmatrix}$	$\begin{smallmatrix}1&0\\0&-1\end{smallmatrix}$	$\begin{smallmatrix}-1&0\\0&1\end{smallmatrix}$	$\begin{smallmatrix}0&1\\1&0\end{smallmatrix}$	$\begin{smallmatrix}0&-1\\-1&0\end{smallmatrix}$	$\begin{smallmatrix}0&-1\\1&0\end{smallmatrix}$	$\begin{smallmatrix}0&1\\-1&0\end{smallmatrix}$	$\begin{smallmatrix}1&0\\0&1\end{smallmatrix}$	$\begin{smallmatrix}-1&0\\0&-1\end{smallmatrix}$	$\begin{smallmatrix}1&0\\0&-1\end{smallmatrix}$	$\begin{smallmatrix}-1&0\\0&1\end{smallmatrix}$	$\begin{smallmatrix}0&1\\1&0\end{smallmatrix}$	$\begin{smallmatrix}0&-1\\-1&0\end{smallmatrix}$	$\begin{smallmatrix}0&-1\\1&0\end{smallmatrix}$	$\begin{smallmatrix}0&1\\-1&0\end{smallmatrix}$

Table 6 shows that there are 10 *irreps*, eight of them are 1D (Γ_{1-8}) and two are 2D (Γ_{9-10}). The application of orthogonality rules leads easily to the subsequent decomposition of Γ:

$$\Gamma_m \{\mathbf{k} = (0, 0, 0)\} = \Gamma_2 \oplus \Gamma_3 \oplus \Gamma_4 \oplus \Gamma_6 \oplus \Gamma_7 \oplus \Gamma_8 \oplus \Gamma_9 \oplus 2\,\Gamma_{10}$$
$$\Gamma_m \{\mathbf{k} = (0, 0, 1/2)\} = \Gamma_1 \oplus \Gamma_2 \oplus \Gamma_3 \oplus \Gamma_5 \oplus \Gamma_6 \oplus \Gamma_7 \oplus 2\Gamma_9 \oplus \Gamma_{10}$$

Figure 9 shows the magnetic structures (basis vectors of Γ_i) which have been obtained, by

Figure 9: Magnetic structures for Γ_2, Γ_3, Γ_4, Γ_6, Γ_7 and Γ_8 *irreps* corresponding to *P4/mbm* space-group for propagation vector $\mathbf{k} = (0, 0, 0)$ and ($4h$) Wyckoff position.
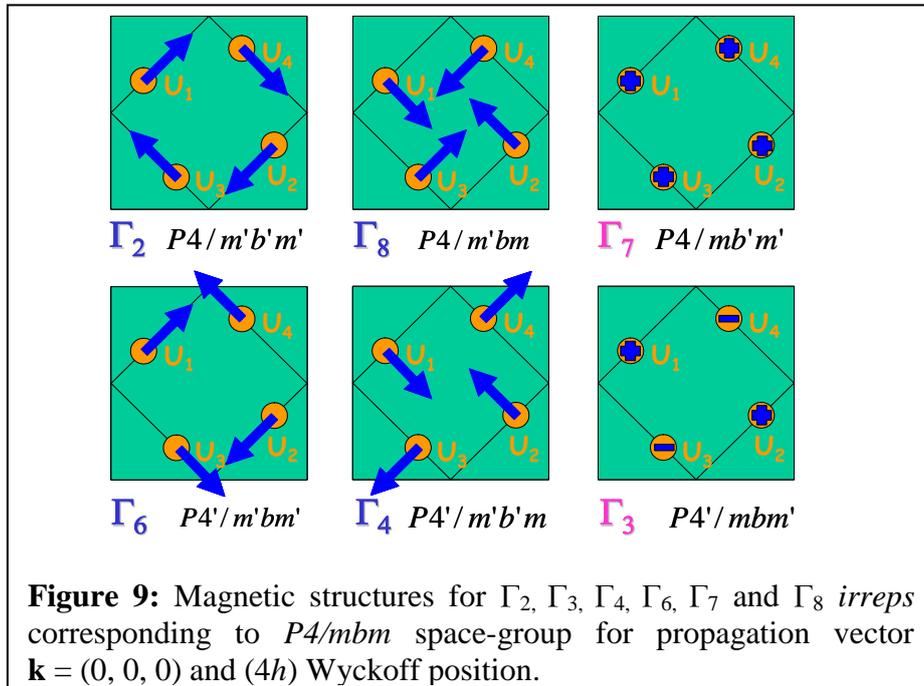
applying the formula (54), for each one of the six 1D *irreps* contained in the Γ_m {$\mathbf{k} = (0, 0, 0)$} decomposition. For all Γ_i, with i = 2, 3, 4, 6, 7 or 8, the associated magnetic structure depends only on a single parameter: the value of the common magnetic moment for U_1 to U_4 atoms.

In Figure 9 we give the symbols of the Shubnikov groups in one-to-one correspondence with the 1D *irreps*. As Γ_m {$\mathbf{k} = (0, 0, 0)$} contains Γ_9 and Γ_{10} they have also to be considered. These *irreps* are both 2D, but are present either once or twice in the Γ decomposition. That means that two and four parameters will be necessary to describe the Γ_9 and Γ_{10} respectively. The general Fourier coefficients obtained from the basis vectors are (adapted from BASIREPS output):

Γ_9
```
SYMM x,y,z                Sk(1): (0,0,u-v)      → S₁:(0,0, α)
SYMM -x+1,-y+1,z          Sk(2): (0,0,-u+v)     → S₂:(0,0,-α)
SYMM -x+1/2,y-1/2,-z+1    Sk(3): (0,0,-u-v)     → S₃:(0,0,-β)
SYMM x+1/2,-y+3/2,-z+1    Sk(4): (0,0,u+v)      → S₄:(0,0, β)
```

Γ_{10}
```
SYMM x,y,z                Sk(1): (u+p,v+w,0)  → S₁:(α,β,0)
SYMM -x+1,-y+1,z          Sk(2): (u+p,v+w,0)  → S₂:(α,β,0)
SYMM -x+1/2,y-1/2,-z+1    Sk(3): (-u+p,v-w,0) → S₃:(γ,δ,0)
SYMM x+1/2,-y+3/2,-z+1    Sk(4): (-u+p,v-w,0) → S₄:(γ,δ,0)
```

We have performed a change of mixing coefficients (last column) in order to simplify the aspect of the Fourier vectors (magnetic moments). Depending on the selection of coefficients we obtain different invariance symmetries for the magnetic structures. For obtaining constant moment magnetic structures the coefficients should be constrained as follows: (Γ_9) $\alpha = \pm\beta$, (Γ_{10}) ($\alpha = \pm\gamma$ and $\beta = \pm\delta$) or ($\alpha = \pm\delta$ and $\beta = \pm\gamma$).

Figure 10a shows a scheme of the U_2Pd_2In magnetic structure, as deduced from symmetry analysis and Neutron Powder Diffraction (NPD) experiments (see Figure 10b). This magnetic structure is characteristic of the *irrep* Γ_6 (*P4'/m'bm'*), with $m_U = 1.55$ μ_B at T=1.4K [33].
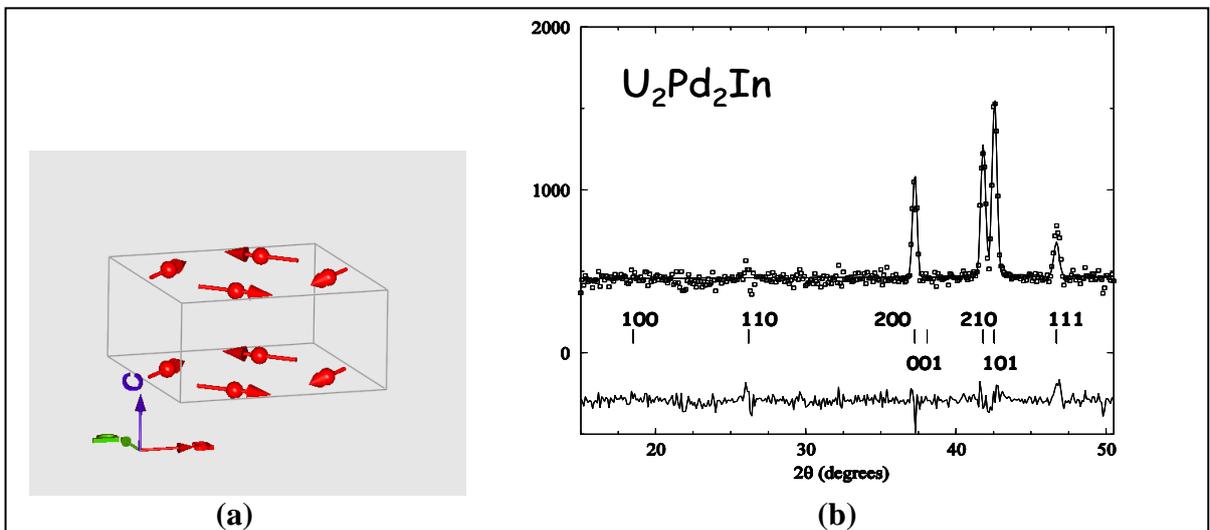
Figure 10: U_2Pd_2In: magnetic structure ($T<T_N$) and difference NPD data [T=1.5K]-[$T>T_N$] collected on G4.1 at LLB, $\lambda = 2.425$Å. Only the magnetic contribution to NPD at T=1.5K data is shown in the figure.

A similar analysis can be done for $\Gamma_m\{\mathbf{k} = (0, 0, 1/2)\}$. We have already shown three different cases of magnetic structures corresponding to this case. The magnetic structures correspond to the following irreps of $\mathbf{G_{k = (0, 0, 1/2)}}$.

Figure 8b: Γ_7 ($P_{2c}4/mb'm'$) : $\mathbf{S}_1 = (m, m, 0)$; $\mathbf{S}_2 = (-m, -m, 0)$; $\mathbf{S}_3 = (-m, m, 0)$, $\mathbf{S}_4 = (m, -m, 0)$

Figure 8c: Γ_2 ($P_{2c}4/m'b'm'$): $\mathbf{S}_1 = (0, 0, m)$; $\mathbf{S}_2 = (0, 0, m)$; $\mathbf{S}_3 = (0, 0, m)$; $\mathbf{S}_4 = (0, 0, m)$

Figure 8d: Γ_6 ($P_{2c}4'/m'bm'$): $\mathbf{S}_1 = (0, 0, m)$; $\mathbf{S}_2 = (0, 0, m)$; $\mathbf{S}_3 = (0, 0, -m)$; $\mathbf{S}_4 = (0, 0, -m)$

This can be easily seen by inspecting the output of the BASIREPS program.

11. Examples of incommensurate magnetic structures.

In this section we provide two examples of simple incommensurate magnetic structures. The first one is a simple sinusoidal structure (single propagation vector, Ce_2Pd_2Sn, from the R_2T_2X family discussed in subsection 10.4) and the second one a conical structure ($DyMn_6Ge_6$ with two propagation vectors belonging to different stars) close to a commensurate superstructure.

11.1: *Incommensurate* $\mathbf{k} = (\delta, 0, 0)$ *magnetic structure of* Ce_2Pd_2Sn

Experimental studies on Ce_2Pd_2Sn magnetic properties [36] are presented in Figures 11-14.

| **Figure 11**: Ce_2Pd_2Sn, magnetic susceptibility $\chi(T)$. | **Figure 12**: Ce_2Pd_2Sn, NPD data (G4.1 - $\lambda = 2.425$Å), at T=1.5K (T<T_C), T=4K (T_C<T<T_N) and T=10K (T>T_N). |

| **Figure 13**: Ce_2Pd_2Sn, NPD data *vs. T*, in [39°, 44°] 2θ-range: (210) Bragg peak and $(210)^-$, $(120)^-$, $(120)^+$, $(210)^+$ magnetic satellites, $\mathbf{k} = (\delta\ 0\ 0)$. | **Figure 14:** Ce_2Pd_2Sn, propagation vector ($\delta\ 0\ 0$): $\delta=k_x$ *vs. T*. Coexistence of commensurate ($\delta = 0$) and incommensurate ($\delta \neq 0$) propagation vectors observed around 3K. |
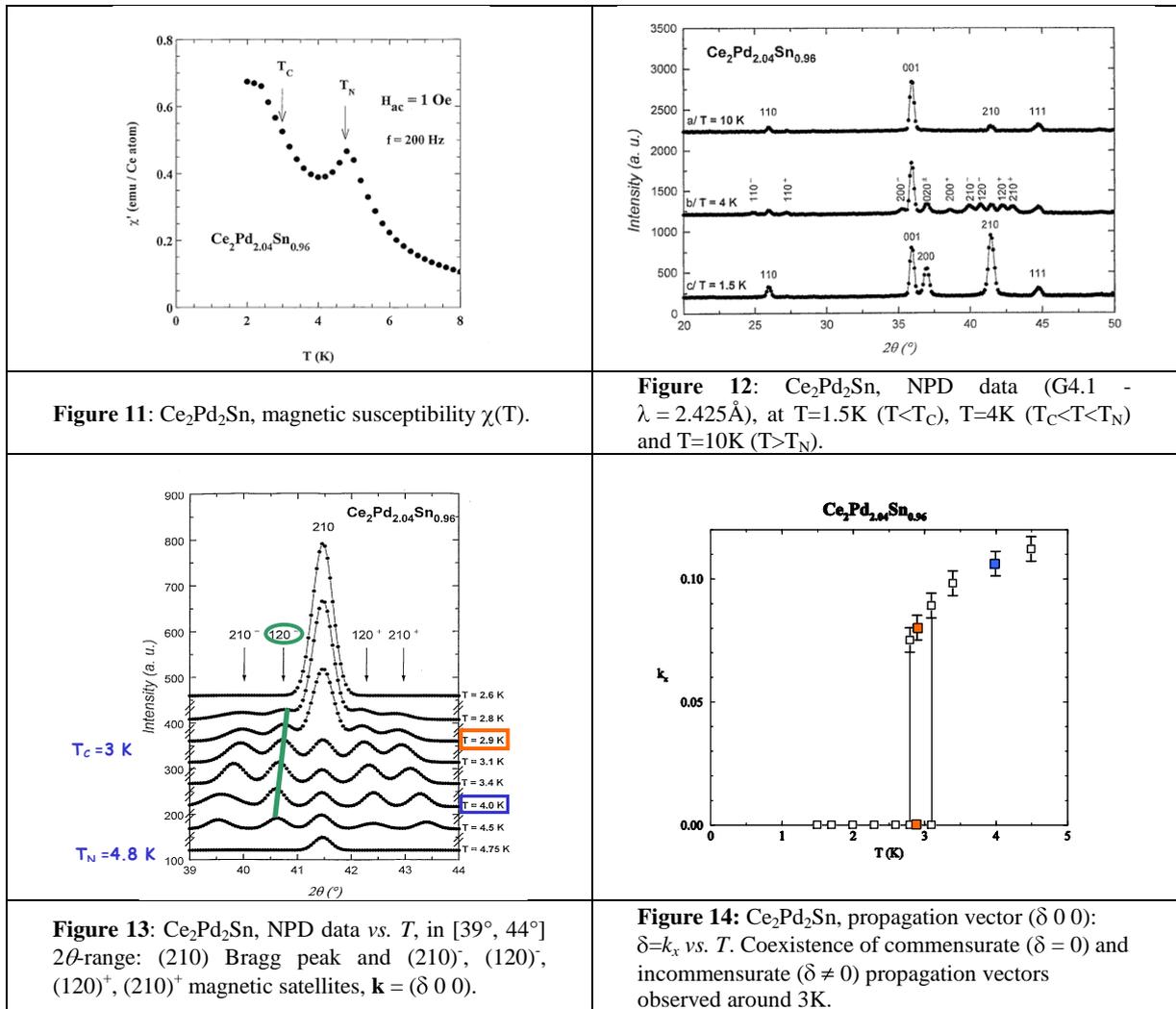
From magnetic susceptibility measurements (χ(T), Figure 11) the existence of two successive magnetic transitions is deduced, at around $T_N = 4.8(2)$ K (antiferromagnetic transition) and $T_C = 3.0(2)$ K (ferromagnetic transition) respectively.

Characteristic NPD data are shown in Figures 12 and 13. Below T = 2.6 K (Figure 12c), the Ce_2Pd_2Sn magnetic structure is ferromagnetic with $\mathbf{k} = (0\ 0\ 0)$ and Ce^{3+} magnetic moments parallel to the tetragonal c-axis. In the intermediate [3.4 K - 4.75 K] T-range, NPD data (Figures 12b and 13) are associated with Ce^{3+} magnetic moments parallel to $\mathbf{c}$-axis. The experimental NPD data can be fit with a very simple magnetic structure in which the moments of the Ce atoms are described as: $\mathbf{m}_{lj} = \mathbf{m}_{Ce}\ cos\{2\pi\mathbf{k}\ (\mathbf{R}_l + \mathbf{r}_j)\}$, with $\mathbf{k} = (\delta, 0, 0)$, δ being T-dependent (see Figure 14) and $\mathbf{m}_{Ce}$ the common amplitude of magnetic moments that is directed along the $\mathbf{c}$-axis.

Whatever the case of an incommensurate propagation vector $\mathbf{k}$, the little group $\mathbf{G_k}$ is no more coincident with $\mathbf{G}$ (*P4/mbm*), but a sub-group of $\mathbf{G}$. For $\mathbf{k} = \delta\ \mathbf{a^*}$, as observed for Ce_2Pd_2Sn in the intermediate T-range, this subgroup includes only four symmetry operators, SYMM(1), SYMM(4), SYMM(10) and SYMM(11), according to Table 4. The list of $\mathbf{G_k}$ operators are given in Table 7.

Table 7: Coset representatives of the little group $\mathbf{G_k}$= *P2₁am* ($\mathbf{k} = \delta\ \mathbf{a^*}$ in *P4/mbm*)

Symmetry operators of $\mathbf{G_k}$	Symbol (rotational part)	Translation	Location	Kovalev Notation (rotational part)
x, y, z	1			h_1
½+*x*, ½-*y*, -*z*	2_x	[½ 0 0]	(0 ¼ 0)	h_2
x, y, -z	m_z			h_{28}
½+*x*, ½-*y*, *z*	m_y	[½ 0 0]	(0 ¼ 0)	h_{27}

The space group may be decomposed in coset representatives with respect to $\mathbf{G_k}$ (*P2₁am*) as follows:

$$\mathbf{G}=P4/mbm=\mathbf{G_k}+ 2_{1y}\ \mathbf{G_k} + 4^-_z\ \mathbf{G_k} + 4^+_z\ \mathbf{G_k}$$

The full star of the propagation vector is formed by four non-equivalent $\mathbf{k}$-vectors:

$$\{\mathbf{k}\} =\{\ \mathbf{k}_1=(\delta, 0, 0);\ \mathbf{k}_2=(-\delta, 0, 0);\ \mathbf{k}_3=(0, \delta, 0);\ \mathbf{k}_4=(0, -\delta, 0)\}$$

The magnetic moments in Ce_2Pd_2Sn are now still located at the (4*h*) Wyckoff position (Ce^{3+} site). But, in the case of $\mathbf{k} = (\delta, 0, 0)$, with $\delta \neq 0$ and $\delta \neq$½, $\mathbf{G_k}$ now splits their sublattices into two independent sets (orbits): Ce_1 and Ce_4 on one side, and Ce_3 and Ce_2 on the other. No symmetry operation in $\mathbf{G_k}$ allows transforming any Ce atom of one orbit to a Ce atom in the other orbit.

These two orbits, Ob_1 and Ob_2, are listed below:

Ob_1	Ce_1	x_{Ce}	½ + x_{Ce}	½
	Ce_4	½ + x_{Ce}	1 - x_{Ce}	½

Ob_2	Ce_3	½ - x_{Ce}	x_{Ce}	½
	Ce_2	1 - x_{Ce}	½ -x_{Ce}	½

Each of these orbits, Ob_1 and Ob_2, is associated to a 6-dimensional magnetic (reducible) representation of $\mathbf{G_k}$, $\Gamma_m(Ob_1)$ and $\Gamma_m(Ob_2)$ respectively.

The Table 8 lists all the *irreps* of $\mathbf{G_{k\ =\ (\delta,\ 0,\ 0)}}$ group. These *irreps* are no more real, but complex, with α parameter being equal to $\alpha = exp\{2\pi i\,(1/2\,\delta)\} = exp\{\pi i\delta\}$, where $\phi = \pi\delta$ is the phase-shift associated to the [1/2, 0, 0] translation of the symmetry operators.

Table 8: Small representations of the little group $\mathbf{G_k} = P2_1am$.

$\mathbf{G_k}$	1	$\{2_x|1/200\}$	$\{m_z|000\}$	$\{m_y|1/200\}$
Γ_1	1	α	1	α
Γ_2	1	α	-1	$-\alpha$
Γ_3	1	$-\alpha$	1	$-\alpha$
Γ_4	1	$-\alpha$	-1	α

Both $\Gamma_m(Ob_1)$ and $\Gamma_m(Ob_2)$ can be decomposed into $\mathbf{G_{k\ =\ (\delta,\ 0,\ 0)}}$ *irreps*. Whatever the orbit, Ob_1 or Ob_2, we get:

$$\Gamma_m = \Gamma_1 \oplus 2\,\Gamma_2 \oplus \Gamma_3 \oplus 2\,\Gamma_4$$

The Fourier coefficient deduced from the basis vectors are listed in the table below. Similar vectors are obtained for {Ce_1, Ce_4} (orbit Ob_1) and {Ce_3, Ce_2} (orbit Ob_2) sets, but, in principle (no symmetry constraint), with no relationship either between w and *w'* (Γ_1, Γ_3) or {*u, v*} and {*u', v'*} (Γ_2, Γ_4) parameters. We can leave these parameters free in the refinement adding a phase factor between the two orbits. In such a case three free parameters define the magnetic structure for (Γ_1, Γ_3) and five parameters for (Γ_2, Γ_4). However we can consider that the *transition channel* comprises always two arms of the star: ($\mathbf{k}$, $-\mathbf{k}$) and one of the symmetry operators relating $\mathbf{k}$ to $-\mathbf{k}$ transforms the Fourier coefficients of the one orbit to the other using the equation (55) or equations (62).

Γ_m	$\mathbf{S}_k(Ce_1)$	$\mathbf{S}_k(Ce_4)$	$\mathbf{S}_k(Ce_3)$	$\mathbf{S}_k(Ce_2)$
Γ_1	$(0, 0, w)$	$\alpha^*\,(0, 0, -w)$	$(0, 0, w')$	$\alpha^*\,(0, 0, -w')$
Γ_2	$(u, v, 0)$	$\alpha^*\,(u, -v, 0)$	$(u', v', 0)$	$\alpha^*\,(u', -v', 0)$
Γ_3	$(0, 0, w)$	$\alpha^*\,(0, 0, w)$	$(0, 0, w')$	$\alpha^*\,(0, 0, w')$
Γ_4	$(u, v, 0)$	$\alpha^*\,(-u, v, 0)$	$(u', v', 0)$	$\alpha^*\,(-u', v', 0)$

We know that at the lowest temperature the magnetic structure is ferromagnetic with all moments along $\mathbf{c}$. A very simple hypothesis is to consider that a simple sinusoidal wave embraces all magnetic atoms in the unit cell (all are *in phase*) that tends towards the ferromagnetic structure when $\delta \rightarrow 0$. This is verified by the representation Γ_3. The aspect of the Fourier coefficients of both orbits is of the same type and can be related to the relative position of the atoms:

$$\mathbf{S}_k\ Ce_4 = \alpha^*\mathbf{S}_k\ Ce_1 = e^{-2\pi i\frac{1}{2}\delta}\,\mathbf{S}_k\ Ce_1 = e^{-2\pi i\mathbf{k}(\mathbf{r}_4 - \mathbf{r}_1)}\,\mathbf{S}_k\ Ce_1$$

$$\mathbf{S}_k\ Ce_2 = \alpha^*\mathbf{S}_k\ Ce_3 = e^{-2\pi i\frac{1}{2}\delta}\,\mathbf{S}_k\ Ce_3 = e^{-2\pi i\mathbf{k}(\mathbf{r}_2 - \mathbf{r}_3)}\,\mathbf{S}_k\ Ce_3$$

Considering that the Fourier coefficients of the atoms Ce_1 and Ce_3 are related in the same way as the internal relation within the orbits we establish the following constraint:

$$\mathbf{S_k}\ Ce_3 = e^{-2\pi i \mathbf{k}(\mathbf{r_3}-\mathbf{r_1})}\mathbf{S_k}\ Ce_1$$

The relation between primed parameters (Ob2) and unprimed parameters (Ob1) is then given by:
$$p' = e^{-2\pi i \mathbf{k}(\mathbf{r_3}-\mathbf{r_1})}\ p = e^{-2\pi i \delta(1/2-2x_{Ce})}\ p$$

This constraint fixes the phase between the two orbits to $\phi = \delta\,(1/2\text{-}2x_{Ce})$ (in fraction of 2π) and provides the same amplitude of the Fourier coefficients of the two orbits. This constraint is quite well verified experimentally (see Figure 15a) for *irrep* Γ_3.

The Γ_3 Ce$_2$Pd$_2$Sn incommensurate magnetic structure is shown on Figure 16, either within the origin crystal unit-cell (a×b×c) [Figure 16a] or within (10a×b×c) volume [Figure 16b].
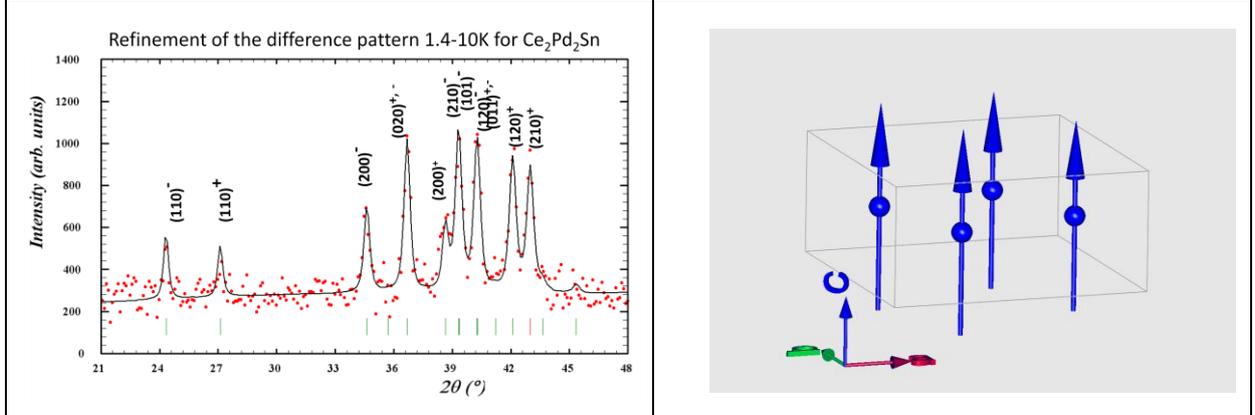
Figure 15. Portion of the pure magnetic NPD pattern of Ce$_2$Pd$_2$Sn refined with the model corresponding to Γ_3	**Figure 16a.** Ce$_2$Pd$_2$Sn, **k** = (0.105, 0, 0) magnetic structure
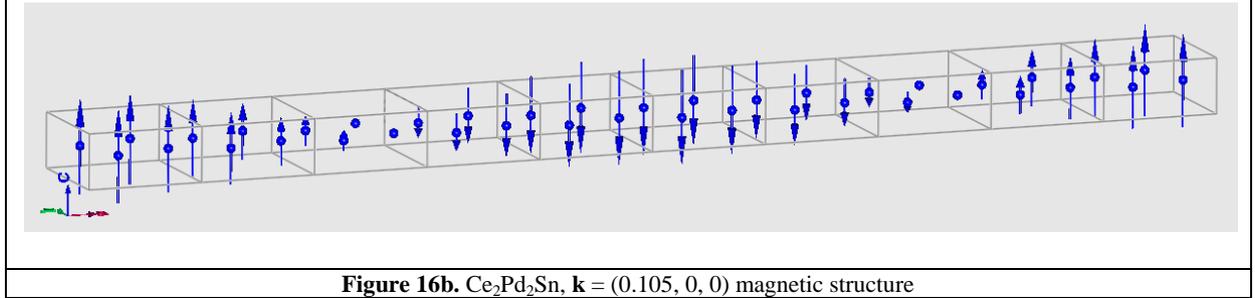
Figure 16b. Ce$_2$Pd$_2$Sn, **k** = (0.105, 0, 0) magnetic structure

We can rewrite the Fourier coefficients for all representations and the four atoms multiplying by the phase factor $exp(-2\pi i\,\mathbf{kr_1})$, this does not change the calculated diffraction pattern, and then we obtain, for instance, for *irrep* Γ_3:
$$\mathbf{S_k}\ Ce_j = 0,0,w\ e^{-2\pi i \mathbf{k}\,\mathbf{r_j}}$$

We can see, looking at the Fourier series defined as in (34) and (34'), that:

$$\mathbf{m}_{lj} = \sum_{\mathbf{k},-\mathbf{k}}\mathbf{S}_{\mathbf{k}j}e^{-2\pi i \mathbf{k}\mathbf{R_l}} = \sum_{\mathbf{k},-\mathbf{k}}(0,0,w)e^{-2\pi i \mathbf{k}\mathbf{r_j}}e^{-2\pi i \mathbf{k}\mathbf{R_l}} = \sum_{\mathbf{k},-\mathbf{k}}(0,0,w)exp(-2\pi i \mathbf{k}(\mathbf{R_l}+\mathbf{r_j}))$$

$$\mathbf{m}_{lj} = 2\,(0,0,w)\cos\{2\pi\mathbf{k}(\mathbf{R_l}+\mathbf{r_j})\} = (0,0,m_{Ce})\cos\{2\pi\mathbf{k}(\mathbf{R_l}+\mathbf{r_j})\}$$

The Fourier coefficients $\mathbf{T_{kj}} = (0, 0, w)$ are all identical and real in *irrep* Γ_3.
Experimentally, the refinement of the phase factor between the two orbits constraining the amplitudes to be the same (two free parameters) gives ϕ=0.015(6) to be compared with the expected value ϕ=$\delta\,(1/2\text{-}2x_{Ce})$=0.105 ×(0.5-2×0.1781)= 0.0151. The refined value of the amplitude is m_{Ce} = 1.70(5) μ_B at T = 4K.

11.2: *A conical magnetic structure:* DyMn$_6$Ge$_6$ *with* $\mathbf{k}_1 = (0, 0, 0)$ *and* $\mathbf{k}_2 = (0, 0, \delta)$

The compound DyMn$_6$Ge$_6$ crystallizes in the well known HfFe$_6$Ge$_6$ structure [37]: space group *P6/mmm*, unit cell parameters $a \approx 5.21$ Å, $c \approx 8.15$ Å. All the compounds RMn$_6$Ge$_6$ (R: rare earth) have a macroscopic ferrimagnetic behaviour, however their magnetic structures are quite complex and vary with temperature. The magnetic atoms occupy the Wyckoff positions R (1*a*) and Mn (6*i*) with the representative first positions (0, 0, 0) and (1/2, 0, *z*), respectively. The Mn atoms form two Kagomé planes perpendicular to the **c**-axis (Figure 17). We present here the low temperature magnetic structure of DyMn$_6$Ge$_6$ [38] (see also Table 3 of reference [39] for corrected parameters) that is an example of the coexistence of two propagation vectors $\mathbf{k}_1 = (0, 0, 0)$ and $\mathbf{k}_2 = (0, 0, \delta)$ (Figure 18) and we will see that the magnetic structure can be described as a double *conical* structure. The value of $\delta \approx 0.1651(4)$ is quite close to a commensurate rational value ($\delta = 1/6$), so that the structure may also be described with an approximate Shubnikov group.

Figure 17: Magnetic sublattices in DyMn$_6$Ge$_6$, showing the Kagomé planes of Mn atoms and the Dy atom at the origin of the unit cell

The symmetry analysis using the representation theory and the program BASIREPS [24], provides immediately the little groups $\mathbf{G}_{\mathbf{k}_1}$ = *P6/mmm* and $\mathbf{G}_{\mathbf{k}_2}$ = *P6mm*. The numbering of *irreps* is that provided by the program. We do not write explicitly all the *irreps* in the present case. There are eight 1D *irreps* and four 2D *irreps* for $\mathbf{G}_{\mathbf{k}1}$ and four 1D *irreps* and two 2D *irreps* for $\mathbf{G}_{\mathbf{k}2}$. From the analysis of the magnetic representation for both sites:

$$\Gamma_{m,\mathbf{k}_1=(0,0,0)} \ \text{Dy-}1a \ \ = \Gamma_3 \oplus \Gamma_{12}$$

$$\Gamma_{m,\mathbf{k}_1=(0,0,0)} \ \text{Mn-}6i \ \ = \Gamma_2 \oplus \Gamma_3 \oplus \Gamma_5 \oplus \Gamma_6 \oplus \Gamma_7 \oplus \Gamma_8 \oplus \Gamma_9 \oplus \Gamma_{10} \oplus 2\Gamma_{11} \oplus 2\Gamma_{12}$$

it is clear that a strong candidate to explain the ferrimagnetic component observed macroscopically is the representation Γ_3, for which the magnetic moments are aligned along the **c**-axis for both sites: Dy(0, 0, $m_{z\text{Dy}}$) and Mn$_{j=1,\ldots6}$(0, 0, $m_{z\text{Mn}}$). This representation should be combined with the appropriate representation(s) corresponding to $\mathbf{k}_2 = (0, 0, \delta)$. The Wyckoff positions of Mn (6*i*) are now split in two orbits of three atoms each. The decomposition of the magnetic representation in small *irreps* is as follows:

$$\Gamma_{m,\mathbf{k}_2=(0,0,\delta)} \ \text{Dy-}1a \ \ = \Gamma_2 \oplus \Gamma_6$$

$$\Gamma_{m,\mathbf{k}_2=(0,0,\delta)} \ \text{Mn-}6i \ \ = \Gamma_2 \oplus \Gamma_3 \oplus \Gamma_4 \oplus \Gamma_5 \oplus 2\Gamma_6$$

Figure 18: Powder diffraction pattern of DyMn$_6$Ge$_6$ at 11K taken at DMC (PSI) with $\lambda = 2.425$Å. The first propagation vector is $\mathbf{k}_1 = (0, 0, 0)$ and the second one is $\mathbf{k}_2 = (0, 0, \delta)$ with $\delta \approx 0.1651(4)$, quite close to $\delta = 1/6$. The calculated nuclear diffraction pattern is shown as a continuous line.
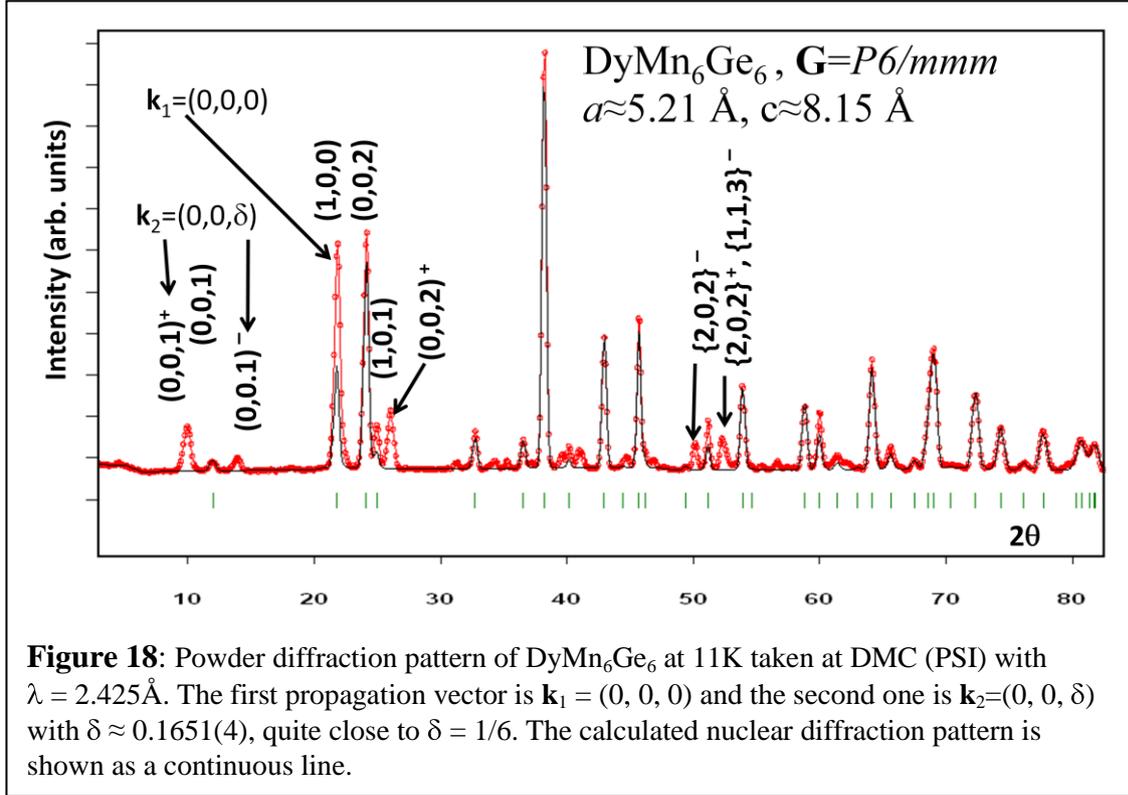
The representation Γ_2 for Dy provides a basis function with Fourier components along **c**:

$$\mathbf{S}_{\mathbf{k}_2,\text{Dy},\Gamma_2} = (0,0,u)$$

The Fourier coefficients for Mn atoms are all of the same type (along the **c**-axis) as those for Dy atom. The representation Γ_2 for $\mathbf{k}_2$ combined with the representation Γ_3 for $\mathbf{k}_1$ will give a collinear magnetic structure with fluctuating magnetic moments along the **c**-axis. This can be immediately rejected by comparing with experimental data.

The special position occupied by Dy atoms (a simple Bravais lattice) induces strong constraints for the possible magnetic models for describing the whole magnetic structure. The only possibility is to consider the representation Γ_6 (2D *irrep*) that gives a Fourier component for Dy that is within the *ab* plane:

$$\mathbf{S}_{\mathbf{k}_2,\text{Dy},\Gamma_2} = \left(u,v,0\right) + \frac{i}{\sqrt{3}}\left(u+2v,2u+v,0\right) = \frac{1}{2}\left(\mathbf{R}+i\,\mathbf{I}\right)$$

If we put the restriction of a constant magnetic moment, the real part **R** must be perpendicular to the imaginary part **I** (e.g. $\mathbf{R}\cdot\mathbf{I}=0$). This is obtained using real values for u and v and by putting $v=0$ (or $u=0$), so:

$$\mathbf{S}_{\mathbf{k}_2,\text{Dy},\Gamma_2} = \left(u,0,0\right) + \frac{i}{\sqrt{3}}\left(u,2u,0\right)$$

It is very easy to verify that $(u, 0, 0)$ is perpendicular to $(u, 2u, 0)$ in the hexagonal reference frame. The magnetic structure for the Dy atoms corresponds in this case (if we combine with Γ_3 for $\mathbf{k}_1$) to a conical structure, described by the expression:

$$\mathbf{m}_l(\mathrm{Dy}) = \sum_{\mathbf{k}_1,\mathbf{k}_2,-\mathbf{k}_2} \mathbf{S}_\mathbf{k}(\mathrm{Dy}) e^{-2\pi i \mathbf{k}\mathbf{R}_l} = (0,\ 0, m_{z\mathrm{Dy}}) +$$

$$2\,(u,0,0)\cos\{2\pi\mathbf{k}_2\mathbf{R}_l\} + \frac{2}{\sqrt{3}}(u,2u,0)\sin\{2\pi\mathbf{k}_2\mathbf{R}_l\}$$

The vectors are given with respect to the unitary frame along the axes of the conventional cell. If we adopt a Cartesian frame with $\mathbf{x}$ along $\mathbf{a}$, $\mathbf{y}$ in the ab plane and $\mathbf{z}$ along $\mathbf{c}$, the Fourier coefficient for Dy is then: $\mathbf{S}_{\mathbf{k}_2,\mathrm{Dy},\Gamma_2} = 1/2\,m_{\mathrm{Dy}}(\mathbf{x}+i\,\mathbf{y})$, with $m_{\mathrm{Dy}}=2u$ the amplitude of the helical moment of Dy. The equation can be simplified as (remember that $\mathbf{k}_2 = (0, 0, \delta)$):

$$\mathbf{m}_l(\mathrm{Dy}) = \sum_{\mathbf{k}_1,\mathbf{k}_2,-\mathbf{k}_2} \mathbf{S}_\mathbf{k}(\mathrm{Dy}) e^{-2\pi i \mathbf{k}\mathbf{R}_l} = m_{\mathrm{Dy}}\cos(2\pi\delta l_3)\mathbf{x} + m_{\mathrm{Dy}}\sin(2\pi\delta l_3)\mathbf{y} + m_{z\mathrm{Dy}}\mathbf{z}$$

The amplitude of the magnetic moments of the Dy atoms is independent of the particular unit cell of the crystal and is given by:

$$m_l(\mathrm{Dy}) = \sqrt{\mathbf{m}_l.\mathbf{m}_l} = \sqrt{m_{\mathrm{Dy}}^2\cos^2(2\pi\delta l_3) + m_{\mathrm{Dy}}^2\sin^2(2\pi\delta l_3) + m_{z\mathrm{Dy}}^2} = \sqrt{m_{\mathrm{Dy}}^2 + m_{z\mathrm{Dy}}^2}$$

The full cone angle is given by the expression $\beta_{\mathrm{Dy}} = 2\arctan(m_{\mathrm{Dy}}/m_{z\mathrm{Dy}})$. The pitch angle of the helical part is determined by the propagation vector: $\alpha = 2\pi\delta \approx 59.4°$, corresponding nearly to 60°. The conical structure for Dy atoms (only two free parameters) breaks the 6-fold symmetry of the little groups of $\mathbf{k}_1$ and $\mathbf{k}_2$. The only symmetry operator of the original space group that remains in this spin configuration is the centre of symmetry in which the Dy atom is sited. It is then possible that the basis functions of the *irrep* Γ_6 for Mn atoms are not adequate to construct a magnetic model explaining the experimental diffraction pattern. This is effectively the case. The Fourier coefficients of Mn atoms in the Γ_6 *irrep* as obtained from the output of BASIREPS are:

```
SYMM x,y,z                      Atom: Mn1_1     0.5000   0.0000   0.2506
Sk(1): (u-r0.w+r0.p,v+r0.p,0)+i.r1.(-w+p,p,0)

SYMM -y,x-y,z                   Atom: Mn1_2     0.0000   0.5000   0.2506
Sk(2): (r0.v+p,-r0.u+r0.v+w,0)+i.r1.(v,-u+v,0)

SYMM -x+y,-x,z                  Atom: Mn1_3    -0.5000  -0.5000   0.2506
Sk(3): r0.(u-v+w,u+w-p,0)+i.r1.(-u+v-w,-u-w+p,0)

Values of real constants r0, r1...
    r0 =   0.500000 = cos(π/3)    r1 =   0.866026 = sin(π/3)
```

Similar Fourier coefficients are obtained for the other three atoms of the other orbit related by the inversion centre at the origin. The number of free parameters without considering symmetry is: 6 atoms × 6 Fourier components (3 real + 3 imaginary) =36 (see equation (58)). For the given orbit we have up to 8 free parameters ($n_v = 2$, $dim\,\Gamma_6 = 2 \rightarrow 2 \times n_v \times dim\,\Gamma_6 = 8$, see equation (58)) for the most general magnetic model. If we consider the highest symmetry, the atoms of the other orbit have Fourier coefficients constrained to be the same (in-phase) or in anti-phase to those of the orbit given above. If we consider as completely independent the second orbit we have a total of 16 free parameters to handle. The Fourier coefficients can further be simplified if we select particular directions in the representation space (selection of free parameters). For instance, if we consider $p = w = 0$ we have:

$$\mathbf{S_k}(1)=(u, v, 0); \qquad \mathbf{S_k}(2)=(v, v\text{-}u, 0)\, \exp\{i\pi/3\}; \quad \mathbf{S_k}(3)=(u\text{-}v, u, 0)\, \exp\{\text{-}i\pi/3\}$$

A similar set of Fourier coefficients is obtained for $u = v = 0$ and other cases can be found by establishing relations between coefficients. In all cases a magnetic moment configuration reminiscent of the 6-fold axis is obtained but none of them corresponds to the observed diffraction pattern. A reduction of symmetry is necessary for obtaining appropriate Fourier coefficients. An indication on how to handle this problem comes from the observation that in all this kind of compounds the observed Mn-Mn interactions are ferromagnetic. A spin configuration that tries to satisfy the requirement of ferromagnetic interaction implies Fourier coefficients identical for all six Mn atoms in the (6*i*) positions but affected by opposite phase factors

$$\mathbf{S_k}(1, 2, 3)= \tfrac{1}{2}\, m_{\mathrm{Mn}}\, (\mathbf{x} + i\,\mathbf{y})\, \exp\{\text{-}2\pi i\phi\}; \quad \mathbf{S_k}(4,5,6)= \tfrac{1}{2}\, m_{\mathrm{Mn}}\, (\mathbf{x} + i\,\mathbf{y})\, \exp\{2\pi i\phi\}$$

That gives only two free parameters for describing the helical part of the magnetic structure of the Mn atoms. We have used the same Cartesian frame that we discussed for Dy atoms. The magnetic moment of the Mn atoms can be described by the formula:

$$\mathbf{m}_l(\mathrm{Mn}_j) = \sum_{\mathbf{k}_1, \mathbf{k}_2, -\mathbf{k}_2} \mathbf{S_k}(\mathrm{Mn}_j) e^{-2\pi i \mathbf{k}\mathbf{R}_l} = m_{\mathrm{Mn}} \cos\{2\pi(\delta l_3 + \phi_j)\}\mathbf{x} + m_{\mathrm{Mn}} \sin\{2\pi(\delta l_3 + \phi_j)\}\mathbf{y} + m_{z\,\mathrm{Mn}}\mathbf{z}$$

with $\phi_j = \phi$ for $j = 1, 2, 3$ and $\phi_j = \text{-}\phi$ for $j=4, 5, 6$. The basis functions corresponding to the above Fourier coefficients can be obtained by mixing the appropriate representations of the ortho-hexagonal subgroup of *P6/mmm*, *Cmmm* (2**a**+**b**, **b**, **c**) for **k** = (0, 0, δ) and making the constraint of equal moments for the Mn atoms. The use of this model gives a very good agreement with the experimental data. In Figure 19 we present the refinement of the powder diffraction pattern with the discussed model. Pictures of the magnetic structure are also shown.

We give below the relevant part of the input file for FullProf corresponding to the description of the magnetic structure with two propagation vectors, using spherical components for the Fourier coefficients that allow an easy control of the perpendicular constraint of the real and imaginary parts. The total number of free parameters for the magnetic structure is 5.

```
.......
!-------------------------------------------------------------------------------
! Data for PHASE number:  2  ==> Current R_Bragg for Pattern#  1:     4.82
!-------------------------------------------------------------------------------
DyMn6Ge6 magnetic
!
!Nat Dis Mom Pr1 Pr2 Pr3 Jbt Irf Isy Str Furth      ATZ     Nvk Npr More
  6   0   0 0.0 0.0 1.0  -1  -1  -1   0   0          0.000   -2   7  0
!
P 6 m m                 <--Space group symbol for hkl generation
!Nsym Cen Laue MagMat
   3   1  12    1
!
SYMM   x, y, z
MSYM   u, v, w, 0.00
SYMM  -y, x-y, z
MSYM   u, v, w, 0.00
SYMM  -x+y, -x, z
MSYM   u, v, w, 0.00
!
!Atom   Typ  Mag Vek    X        Y        Z        Biso     Occ      Rm      Rphi   Rtheta
!    Im   Iphi   Itheta  beta11  beta22  beta33   MagPh
Dy    JDY3  1 -2  0.00000 0.00000 0.00000 0.00000  0.33333   6.195   0.000  90.000
```

```
                                    0.00      0.00      0.00      0.00      0.00    21.00      0.00      0.00
        6.195   90.000   90.000    0.000     0.000     0.000     0.00000
       21.00      0.00     0.00     0.00      0.00      0.00      0.00
Dy     JDY3   1  1   0.00000  0.00000  0.00000  0.00000   0.33333    3.932     0.000     0.000
                                    0.00      0.00      0.00      0.00      0.00   161.00      0.00      0.00
        0.000    0.000    0.000     0.000     0.000     0.000     0.00000
        0.00      0.00     0.00     0.00      0.00      0.00      0.00
Mn1    MMN2   1 -2   0.50000  0.00000  0.25098  0.00000   1.00000    1.783     0.000    90.000
                                    0.00      0.00    81.00      0.00      0.00    31.00      0.00      0.00
        1.783   90.000   90.000    0.000     0.000     0.000     0.55732
       31.00      0.00     0.00     0.00      0.00      0.00     71.00
Mn1    MMN2   1  1   0.50000  0.00000  0.25098  0.00000   1.00000   -1.149     0.000     0.000
                                    0.00      0.00    81.00      0.00      0.00   171.00      0.00      0.00
        0.000    0.000    0.000     0.000     0.000     0.000     0.00000
        0.00      0.00     0.00     0.00      0.00      0.00      0.00
Mn2    MMN2   1 -2  -0.50000  0.00000 -0.25098  0.00000   1.00000    1.783     0.000    90.000
                                    0.00      0.00   -81.00      0.00      0.00    31.00      0.00      0.00
        1.783   90.000   90.000    0.000     0.000     0.000    -0.55732
       31.00      0.00     0.00     0.00      0.00      0.00    -71.00
Mn2    MMN2   1  1  -0.50000  0.00000 -0.25098  0.00000   1.00000   -1.149     0.000     0.000
                                    0.00      0.00   -81.00      0.00      0.00   171.00      0.00      0.00
        0.000    0.000    0.000     0.000     0.000     0.000     0.00000
        0.00      0.00     0.00     0.00      0.00      0.00      0.00
!-------> Profile Parameters for Pattern #  1
! Scale       Shape1      Bov        Str1       Str2       Str3    Strain-Model
  92.370      0.00000    0.60736    0.00000    0.00000    0.00000       0
   11.00000     0.000     91.000     0.000      0.000      0.000
!     U           V           W           X           Y         GauSiz    LorSiz Size-Model
   1.683215   -1.301411   0.378740   0.000000   0.023638   0.000000   0.000000      0
   61.000     141.000    151.000      0.000     51.000      0.000      0.000
!     a           b           c         alpha       beta       gamma     #Cell Info
   5.208272   5.208272   8.152275   90.000000  90.000000 120.000000
 111.00000   111.00000  131.00000     0.00000    0.00000  111.00000
! Pref1    Pref2       Asy1       Asy2       Asy3       Asy4      S_L        D_L
  1.00000   0.00000    0.00000    0.00000    0.00000    0.00000   0.02495    0.03168
   0.00      0.00       0.00       0.00       0.00       0.00      0.00       0.00
! Propagation vectors:
   0.0000000   0.0000000   0.0000000            Propagation Vector  1
   0.000000    0.000000    0.000000
   0.0000000   0.0000000   0.1648251            Propagation Vector  2
   0.000000    0.000000  121.000000
. . . . . . . . . .
```

The refined values of the magnetic free parameters, and their standard deviations, given in the text and corresponding to the values of the above input file, are the following:

$$\mathbf{k}_1 = (0, 0, 0),$$
$$\mathbf{S}_{\mathbf{k}_1}(\mathrm{Mn}) = -1.149(64)\ \mathbf{z}$$
$$\mathbf{S}_{\mathbf{k}_1}(\mathrm{Dy}) = 3.932(87)\ \mathbf{z}$$

$$\mathbf{k}_2 = (0, 0, \delta),\ \ \delta \approx 0.16483(17),$$
$$\mathbf{S}_{\mathbf{k}_2}(\mathrm{Dy}) = \frac{1}{2}\ 6.195(73)\ \ \mathbf{x} + i\ \mathbf{y}$$
$$\mathbf{S}_{\mathbf{k}_2}(\mathrm{Mn}_{1,2,3}) = \frac{1}{2}\ 1.783(39)\ \ \mathbf{x} + i\ \mathbf{y}\ \ \exp\ -2\pi i\ 0.5573(15)$$
$$\mathbf{S}_{\mathbf{k}_2}(\mathrm{Mn}_{4,5,6}) = \frac{1}{2}\ 1.783(39)\ \ \mathbf{x} + i\ \mathbf{y}\ \ \exp\ 2\pi i\ 0.5573(15)$$

It is easy to see that the magnetic moments of Mn or Dy atoms within a basal plane are all parallel between them (as a result of dominant ferromagnetic Mn-Mn and Dy-Dy interactions). The orientation of Mn moments is nearly antiparallel with that of Dy atoms. The Mn-Dy interactions must be mostly antiferromagnetic and a competition along the **c**-axis between first and second neighbours must be responsible of the incommensurability. The full

cone angle of the Dy sublattice is $\beta_{Dy} = 2 \arctan(6.195/3.932) \approx 115.19°$ and that of the Mn sublattices is $\beta_{Mn} = 2 \arctan(1.783/1.149) \approx 114.40°$, quite close to that of Dy atoms.

Figure 19: Refined powder diffraction pattern of DyMn$_6$Ge$_6$ at 11 K with the model discussed in the text: double conical structure with a pitch angle ($2\pi\delta$) close to 60°. Asterisks signal impurity lines. The scheme of the magnetic structure on the left corresponds to a view along the **c**-axis whereas the right panel has the **c**-axis horizontal.
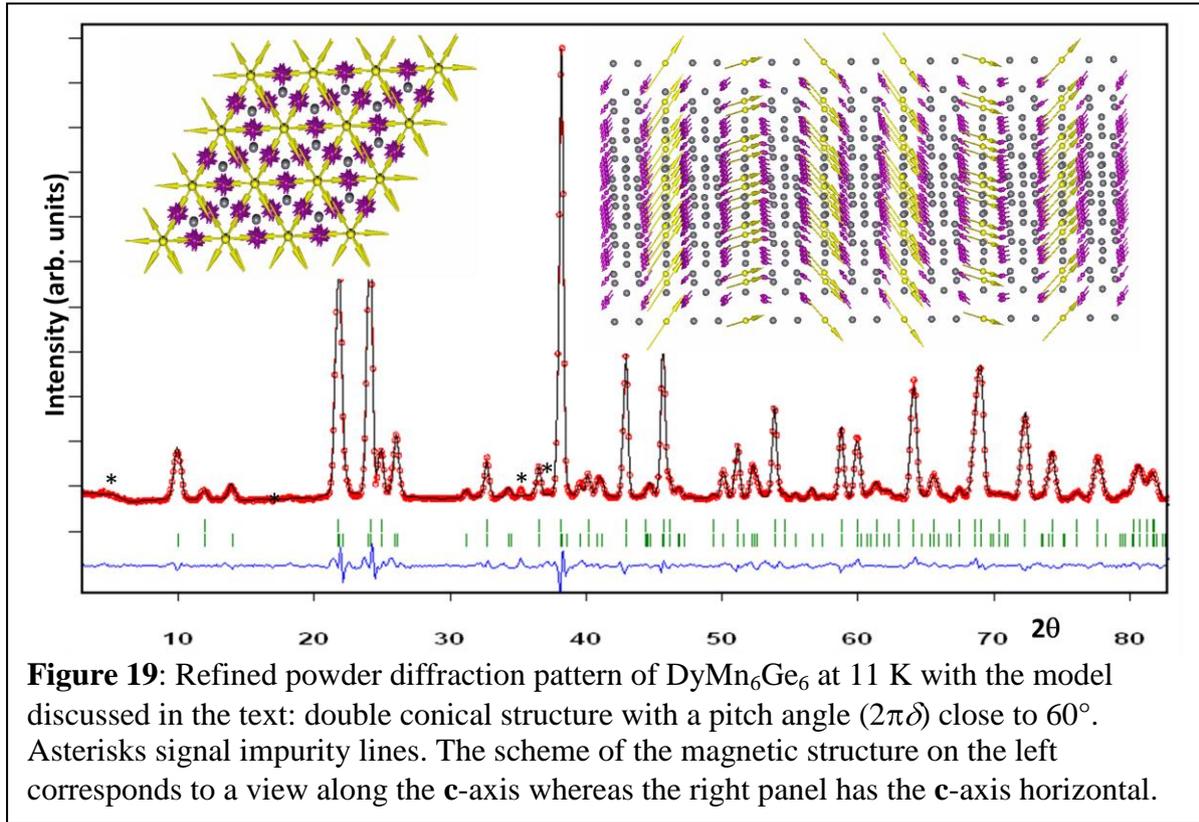
Notice that the refined phase $\phi = 0.5573(15) = ½ + 0.0573(15)$ is not equal to what will be expected from the consideration that a common modulation wave embraces the whole structure $\phi_c = ½ + \mathbf{k}\,\mathbf{r}_{Mn} = ½ + 0.16483 \times 0.25098 = ½ + 0.04137$. The difference $\Delta \approx 0.016$ is higher than three standard deviations. This can be tested using the option JBT=5 that describes conical structures in real space (in terms of magnetic moments, cone angles and phases) using an underline Fourier coefficients of the form given by expression (34') in which the explicit atom positions are part of the phases. If we fix the phase $\Delta = 0$ and refine the data we obtain a reasonable refinement with R$_{Mag}$=5.453% but with significant misfit in the lower angle part of the pattern (see figure 20, top). If we free the parameter we obtain $\Delta = 0.0158(16)$ and a better refinement R$_{Mag}$=4.746 % (see figure 20, bottom).
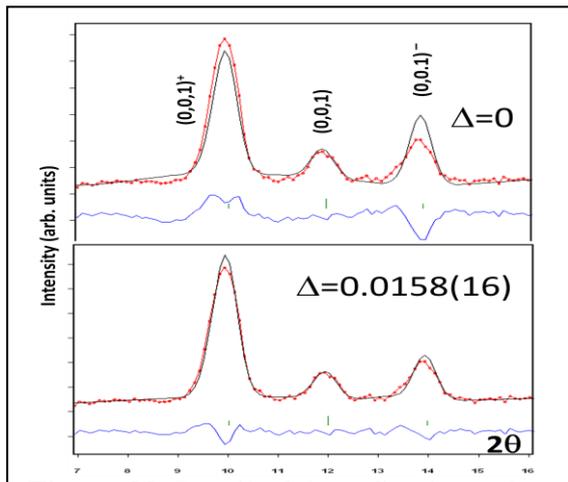
Figure 20: Detail of the refined powder diffraction pattern of DyMn$_6$Ge$_6$ by fixing the phases of Mn atoms to Δ=0 (top) , and leaving free the phases (bottom) using the option JBT=5 in FullProf (see text).

In this kind of refinement (JBT=5), the free parameters are: the value of the moment of Dy and Mn atoms, refined to m(Dy)= 7.34(9)μ_B and m(Mn)=2.11(4) μ_B, respectively, the angle of each magnetic moment with the axis of the cone (half the angle of the cone) and a phase factor per atom. We have constrained the half

angle of the cone of Dy and Mn atoms to have a difference of 180° (same angle but the cone is oriented in the opposite direction). The refined value of the half angle of the cone is 57.5(6)°. The relevant part of input control file for FullProf, using only four free parameters, is given below (all the magnetic atoms must be given explicitly when JBT=5):

```
. . . . . . . . . . . . . . . . .
!-------------------------------------------------------------------------------
! Data for PHASE number:   2  ==> Current R_Bragg for Pattern#  1:     4.75
!-------------------------------------------------------------------------------
DyMn6Ge6 magnetic
!
!Nat Dis Mom Pr1 Pr2 Pr3 Jbt Irf Isy Str Furth     ATZ    Nvk Npr More
   7   0   0 0.0 0.0 1.0   5   0   1   0   0        0.000   -1   7   0
!
P 6 m m                  <--Space group symbol for hkl generation
!Nsym Cen Laue MagMat
   0   1   1   0
!Atom   Typ  Mag Vek    X       Y       Z       Biso    Occ     Mom     Phic    Phase
!  Phi & Theta  of Cone-axis + unused params
Dy     JDY3  1  0   0.00000 0.00000 0.00000 0.00000 1.00000   7.343 -57.527   0.000
                       0.00    0.00    0.00    0.00    0.00 121.00 -151.00    0.00
     0.000   0.000   0.000   0.000   0.000   0.000 0.00000
     0.00    0.00    0.00    0.00    0.00    0.00    0.00
Mn1    MMN2  1  0   0.50000 0.00000 0.25095 0.00000 1.00000   2.113 122.179   0.016
                       0.00    0.00  111.00    0.00    0.00 131.00  151.00  541.00
     0.000   0.000   0.000   0.000   0.000   0.000 0.00000
     0.00    0.00    0.00    0.00    0.00    0.00    0.00
Mn2    MMN2  1  0  -0.50000 0.00000-0.25095 0.00000 1.00000   2.113 122.179  -0.016
                       0.00    0.00 -111.00    0.00    0.00 131.00  151.00 -541.00
     0.000   0.000   0.000   0.000   0.000   0.000 0.00000
     0.00    0.00    0.00    0.00    0.00    0.00    0.00
. . . . . . . . . . . . . . .
Mn6    MMN2  1  0  -0.50000-0.50000-0.25095 0.00000 1.00000   2.113 122.179  -0.016
                       0.00    0.00 -111.00    0.00    0.00 131.00  151.00 -541.00
     0.000   0.000   0.000   0.000   0.000   0.000 0.00000
     0.00    0.00    0.00    0.00    0.00    0.00    0.00
. . . . . . . . . . . . . . .
```

From the refined magnetic structure we can see the presence of approximate invariance symmetry related to the presence of a 6_1 screw axis. If we consider that the propagation vector is rational and exactly equal to $\mathbf{k} = (0, 0, 1/6)$, the crystal and magnetic symmetry can be described by a common Shubnikov group. A simple 6-fold superstructure (**a**, **b**, 6**c**) allows to deduce as subgroups of $\mathbf{G} = P6/mmm$, the groups $\mathbf{H}_1 = P6_122$ and $\mathbf{H}_2 = P6_1$. By inspecting the scheme of the refined magnetic structure it is readily seen that the maximum symmetry corresponds to the Shubnikov group $P6_12'2'$. We know that the propagation vector is changing with temperature above T_t=100K (see reference [38]), however below this temperature the value of δ is locked (within the resolution of the diffraction pattern we can consider that $\delta = 1/6$). With this assumption one can use the Shubnikov group $P6_12'2'$ (**a**, **b**, 6**c**) for describing the symmetry. However if we start from this assumption from the beginning we have to face the fact that in this group we have a single site for Dy atoms and three sites for Mn atoms, in principle with independent values of their magnetic moments. The number of magnetic free parameters in $P6_12'2'$ (**a**, **b**, 6**c**) is 2 components for Dy atoms, which are in a special position with representative magnetic moment (u, $2u$, w), plus three components for each Mn site (3×3=9). So, there are 11 free parameters. I we assume the same value of the magnetic moment for all Mn sites and the ferromagnetic coupling within planes we obtain a model with the same number of free parameters as that of the representation analysis: 2 parameters for Dy and 3 parameters describing all Mn atoms. These constraints are not imposed by symmetry, but it is a physically reasonable assumption. On the other hand, the Shubnikov analysis allows exploring the associated distortions of the nuclear structure by varying the positional parameters of the nuclear structure. We have not used this option here because the Q-range is limited. We have performed a refinement using FullProf in the

Shubnikov group approach (JBT = 10) with only five free magnetic parameters and obtained a comparable refinement (see figure 21).

Figure 21: Refined powder diffraction pattern of $DyMn_6Ge_6$ at 11 K with the model based in the Shubnikov group $P6_12'2'$. In this approach we have constrained the magnetic moments of the Mn atoms in the same basal plane (same z-coordinate) to be ferromagnetically coupled. The obtained values, in μ_B, of the magnetic moment components in the hexagonal unitary frame for Dy are: $\mathbf{m}(Dy) = (3.57(4), 7.14(8), 3.92(9))$; and for those of Mn atoms: $\mathbf{m}(Mn) = (-0.32(2), -1.92(4), -1.13(6))$.

The complete part of input control file for FullProf of a mixed nuclear and magnetic phase is given below:

```
!-------------------------------------------------------------------------------
! Data for PHASE number:   1  ==> Current R_Bragg for Pattern#  1:     3.16
!-------------------------------------------------------------------------------
DyMn6Ge6
!
!Nat Dis Ang Pr1 Pr2 Pr3 Jbt Irf Isy Str Furth      ATZ     Nvk Npr More
   8   0   0 0.0 0.0 1.0  10   0   0   0   0      5566.008    0   7   0
!
P 61 2 2               Magnetic symmetry below
! Time Reversal Operations on Crystal Space Group
  1  1  1  1  1  1 -1 -1 -1 -1 -1 -1
!Atom   Typ      Mag Vek     X          Y          Z        Biso       Occ     N_type  Spc/Fftype /Line
below:Codes
!     Rx       Ry        Rz        Ix        Iy        Iz      MagPh  / Line below:Codes
!     beta11   beta22    beta33    beta12    beta13    beta23  / Line below:Codes
Dy    JDY3      1  0   0.00000    0.00000    0.00000    0.00000    0.50000    1     0
                          0.00       0.00       0.00       0.00       0.00
      3.56539  7.13079   3.92108   0.00000    0.00000    0.00000    0.00000 <-MagPar
     11.00    12.00     21.00      0.00       0.00       0.00       0.00
Mn1   MMN2      1  0   0.50000    0.00000    0.04181    0.00000    1.00000    1     0
                          0.00       0.00      71.00       0.00       0.00
     -0.32364 -1.91453  -1.13012   0.00000    0.00000    0.00000    0.00000 <-MagPar
     31.00    41.00     51.00      0.00       0.00       0.00       0.00
Mn2   MMN2      1  0   0.00000    0.50000    0.04181    0.00000    1.00000    1     0
                          0.00       0.00      71.00       0.00       0.00
     -0.32364 -1.91453  -1.13012   0.00000    0.00000    0.00000    0.00000 <-MagPar
     31.00    41.00     51.00      0.00       0.00       0.00       0.00
Mn3   MMN2      1  0   0.50000    0.50000    0.04181    0.00000    1.00000    1     0
                          0.00       0.00      71.00       0.00       0.00
     -0.32364 -1.91453  -1.13012   0.00000    0.00000    0.00000    0.00000 <-MagPar
     31.00    41.00     51.00      0.00       0.00       0.00       0.00
```

```
Ge1     GE          0  0    0.33333   0.66667   0.08333   0.00000   0.50000    0    0
                             0.00      0.00      0.00      0.00      0.00
Ge12    GE          0  0    0.66667   0.33333   0.08333   0.00000   0.50000    0    0
                             0.00      0.00      0.00      0.00      0.00
Ge2     GE          0  0    0.33333   0.66667   0.00000   0.00000   1.00000    0    0
                             0.00      0.00      0.00      0.00      0.00
Ge3     GE          0  0    0.00000   0.00000   0.05760   0.00000   1.00000    0    0
                             0.00      0.00      81.00     0.00      0.00
!-------> Profile Parameters for Pattern #  1
! Scale       Shape1      Bov       Str1      Str2      Str3    Strain-Model
  2.5702      0.00000    0.60647   0.00000   0.00000   0.00000       0
    61.00000    0.000    101.000    0.000     0.000     0.000
!     U         V          W          X          Y        GauSiz   LorSiz Size-Model
  1.688325  -1.307224   0.379423   0.000000   0.024737  0.000000  0.000000    0
    131.000   141.000    151.000     0.000    171.000     0.000     0.000
!     a         b          c         alpha      beta      gamma       #Cell Info
  5.208386   5.208386   48.914703  90.000000  90.000000 120.000000
   111.00000  111.00000  121.00000   0.00000    0.00000  111.00000
! Pref1    Pref2      Asy1      Asy2      Asy3      Asy4       S_L       D_L
  1.00000  0.00000   0.00000   0.00000   0.00000   0.00000   0.02495   0.03168
    0.00     0.00      0.00      0.00      0.00      0.00      0.00      0.00
```

12.   Final remarks.

We have presented in these notes the basics of the representational analysis of magnetic structures using several examples that illustrate the problems one can encounter when dealing with the analysis of powder neutron diffraction data. These notes do not contain a detailed description of the methods used for determining magnetic structures. The user may find in the web site of the FullProf Suite [28] a series of examples and tutorials that can be followed together with the present notes in order to get a deeper insight into the representational method. We hope, in any case, that the readers will find the presented examples useful for their own research.

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